

1998 Science, Technology, and Technical Support Activities

Self-Assessment

Environment, Safety, and Health (ESH) Division
Los Alamos National Laboratory
Los Alamos, New Mexico

June 1998

*Technology Development, Evaluation,
and Application (TDEA)*

FY 1997 Progress Report

Environment, Safety, and Health (ESH) Division



Los Alamos
NATIONAL LABORATORY

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Abstract

This progress report presents the results of 10 projects funded in FY97 by the Technology Development, Evaluation, and Application (TDEA) Committee of the Environment, Safety, and Health Division. Two of the projects are new for this year and eight continue from FY96. As a result of their TDEA-funded projects, investigators have published 37 papers in professional journals, proceedings, or Los Alamos reports and presented their work at professional meetings. Supplemental funds and in-kind contributions, such as staff time, instrument use, and work space were also provided to the TDEA-funded projects by organizations external to ESH Division. Products generated from the projects funded in FY97 included implementation of radiation worker dosimetric monitoring systems (two); evaluation and validation of cost-effective animal-tracking systems for environmental studies (two); evaluation of personal protective equipment (two); and development of a method for optimal placement of continuous air monitors in the workplace.

The previous reports in this series, unclassified, are LA-13191-PR and LA-13264-PR

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Los Alamos
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*Technology Development, Evaluation,
and Application (TDEA) FY 1997 Progress Report*

Environment, Safety, and Health (ESH) Division

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Introduction

The public expects that the Los Alamos National Laboratory (LANL, Laboratory) will operate in a manner that prevents negative impacts to the environment and protects the safety and health of its employees and the public. To achieve this goal within budget, the Department of Energy (DOE) and the Laboratory must develop new and improved environment, safety, and health (ES&H) technologies and implement innovative, more cost-effective ES&H approaches to operations.

In FY95, the Environment, Safety, and Health (ESH) Division allocated \$300K budget (<1% of its operating budget) to initiate a Technology Development, Evaluation, and Application (TDEA) program. The purpose of this unique program is to test and develop technologies that solve Laboratory ES&H problems and improve the safety of Laboratory operations. Funding for the first year supported six months of work (see LA-13191PR); FY96 was the first full year of the program (see LA-13264-PR) and ESH Division increased the allocation for TDEA-funded projects to \$400K. The acknowledged success of the program led in FY97 to a budget of \$447K, an increase of \$47K.

The TDEA Committee maintains the following program priorities.

- **Improve ES&H protection to Laboratory workers and the public.**

- **Support Laboratory mission objectives.** The first sentence in the Laboratory's mission statement says, "Los Alamos' central mission is reducing the danger of nuclear weapons and nuclear materials worldwide." TDEA projects develop instrumentation and methods to improve on the health and safety of Laboratory workers who directly support the central LANL mission.

- **Respond and build on the unique expertise of the Laboratory and Laboratory requirements.** For example, investigators for TDEA-funded

projects have applied particle physics measurements technology in developing new neutron dosimetry methods; applied sophisticated analytical chemistry techniques to bioassay sample measurement methods; used computer modeling to establish a protocol for efficient placement of continuous air monitors (CAMs) in plutonium facilities.

- **Achieve success within three years.**

Success is demonstrated by the interest, participation, and contributions of the the Laboratory's operating personnel in TDEA-funded projects; by interest from organizations external to the Laboratory; and by the annual increase in the number of papers and presentations prepared for professional journals and meetings.

- **Find potential to transfer technologies to other DOE sites.** Information and methods generated by TDEA-funded projects are transferable to other DOE organizations with the same ES&H concerns. Examples include CAM placement methods, neutron dosimetry methods, bioassay analysis methods, and results from evaluations of protective equipment.

The TDEA Committee assigned priority status to four ES&H areas: dosimetry, instrumentation, monitoring, and neutron measurements. Each area presents an opportunity to improve ES&H performance and further benefit the Laboratory by solving technically complex problems. In FY95, the committee funded five projects and in FY96, seven. In FY97, an increased allocation funded 10 projects, eight of which had been originally funded in past years and two of which were new. In FY97, investigators completed seven projects that resulted in successful technology transfer.

FY97 Projects

- Development and Evaluation of a Radio-Frequency Identification System to Measure Time Spent by Medium-Sized Mammals at Contaminated Sites at Los Alamos National Laboratory (LANL)

- Seasonal Movements, Activity Patterns, and Radionuclide Concentrations of Rocky Mountain Elk (*Cervus elaphus nelsoni*) Inhabiting the Pajarito Plateau

- Applications of Thermal Ionization Mass Spectrometry (TIMS) to the Detection of ²³⁹Pu and ²⁴⁰Pu

- High-Energy Neutron Dosimetry and Spectroscopy

- Development and Implementation of the Los Alamos National Laboratory (LANL) Neutron Extremity Dosimeter

- Optimization of Continuous Air-Monitoring (CAM) Instrument Placement

- Resuspension of ²³⁸Pu from Surfaces

- Reusability of Organic Vapor Air-Purifying Respirator Cartridges

- FRHAM-TEX II Cool Suit Material Testing for Water (and therefore, Tritium) Protection

- Polymeric Barrier Monitor to Protect Workers

Publications and presentations.

Publication in professional refereed journals and presentations at professional meetings are indications of the quality of TDEA-funded projects. In FY96, TDEA-funded projects generated 32 publications and presentations. Publications and presentations increased to 37 in FY97.

Cost savings. Although some TDEA-funded investigators projected cost savings to ESH Division programs, others projected cost savings through technologies that would save time, aid decision making and inform better risk management. Examples of TDEA-generated cost savings:

- Tested the effectiveness of new methods that required less human resource for studying animal activity patterns in and around the Laboratory.

- Applied a sensitive analytical method for monitoring worker exposure, thereby reducing the time required to evaluate exposures and easing the decision-making process; and

- Developed model(s) that allow rapid, optimized placement of workplace air-monitoring equipment for worker protection.

New technologies. TDEA-funded projects that result in developing and testing technologies that yield better protection as well as reduced costs are as follows.

- Evaluated mammal tracking systems that vastly improve the quality of gathered information;
- Developed a CAM placement method that improves plutonium worker protection;
- Gathered information about high-energy neutron measurements to evaluate worker radiation doses that has received world-wide attention;
- Developed analytical techniques and methods using TIMs for rapid determination of worker internal radiation doses; and
- Developed and implemented a long-needed system for measuring extremity doses to plutonium workers.

External funding. Eight TDEA-funded projects attracted external funding from other interested organizations:

- The Dual Axis Radiographic Hydrotesting Facility Mitigation Action Plan contributed \$28K to the Seasonal Movements, Activity Patterns, and Radionuclide Concentrations of Rocky Mountain Elk (*Cervus elaphus nelsoni*) Inhabiting the Pajarito Plateau.
- The NMT-9 group at TA-55 contributed staff time, instrumentation, materials, and a work location for resuspension of ²³⁸Pu from Surfaces.
- The Los Alamos Neutron Science Center (LANCE) contributed beam time for six weeks for the High-Energy Neutron Dosimetry and Spectroscopy project. The rate users are normally charged for beam time is \$300 to \$500 per hour.
- The Environmental System and Waste Characterization Group (CST-7) and the Nuclear and Radiochemistry Group contributed staff time, instrumentation,

materials, and a work location to the Applications of Thermal Ionization Mass Spectrometry (TIMS) to the Detection of ²³⁹Pu and ²⁴⁰Pu project.

- The Optimization of Continuous Air-Monitoring (CAM) Instrument Placement project received \$110K from the Capabilities, Measurement, and Improvement project.
- External funding was also received from two organizations external to the Laboratory: FRHAM-TEX II Cool Suit Material Testing for Water (and therefore, Tritium) Protection received \$9K from Hanford.
- The US Army contributed \$45K to the Reusability of Organic Vapor Air-Purifying Respirator Cartridges project.

The success of TDEA-funded projects over the last three years has demonstrated the innovative approaches by Laboratory ES&H professionals to cost-effectively reduce risk from workplace hazards. We encourage application of the products developed to other processes and facilities. Each additional application of these products increases the cost effectiveness of this program.

This report includes summaries of each project. We invite and encourage readers to followup with the principal investigators for additional information.

Environment



Development and Evaluation of a Radio-Frequency Identification System to Measure Time Spent by Medium-Sized Mammals at Contaminated Sites at Los Alamos National Laboratory (LANL)

Seasonal Movements, Activity Patterns, and Radionuclide Concentrations of Radio-Collared Rocky Mountain Elk (Cervus elaphus nelsoni) Inhabiting the Los Alamos National Laboratory (LANL).

Development and Evaluation of a Radio-Frequency Identification System to Measure Time Spent by Medium-Sized Mammals at Contaminated Sites at Los Alamos National Laboratory (LANL)

Principal investigators: Leslie A. Hansen, Rhonda Robinson, Phil Fresquez, Teralene Foxx, John Huchton, Ecology Group (ESH-20)

Funding: FY97, \$83K

Introduction

Radioactive and nonradioactive contamination of soil, vegetation, invertebrates, and small mammals has been confirmed in areas on LANL property (Fresquez et al. 1996a, b; EAREG 1996; Biggs 1995; Fresquez et al. 1995a, b, c; Brooks 1989). Medium-sized mammals such as raccoons (*Procyon lotor*), bobcats (*Felis rufus*), striped skunks (*Mephitis mephitis*), gray foxes (*Urocyon cinereoargenteus*), squirrels, (*Spermophilus* spp., *Sciurus* spp.) and rabbits (*Sylvilagus* spp.) may be exposed to heavy metals, organic compounds, and low-level radionuclides at the Laboratory (Brooks 1989). Medium-sized mammals are important ecosystem components. They are also used as ceremonial animals by Native Americans, provide an economic resource for trappers, and may be used as food. Carnivores are particularly susceptible to detrimental effects from contaminants because of their acidic digestive systems, which increase the bioavailability of some contaminants, and because of the biomagnification of some contaminants in food chains (Petron 1993, Kendall et al. 1990, Oehme 1978, Stickel 1975).

Although LANL annually conducts extensive monitoring of radioactive and nonradioactive contaminant levels in groundwater, surface water, soils, sediments, the atmosphere, and food-stuffs (EAREG 1996), little information is available on the occurrence of contaminants in natural vegetation (Fresquez et al. 1996b, Fresquez 1995c, Wenzel et al. 1987) and herbivores (Biggs 1995, Fresquez et al. 1995a). To date, no data are available on contaminants in medium-sized mammals. Accumulation of contaminants in individual animals and

contaminant transfer along food chains are among the more important processes that must be evaluated if we are to predict contaminant effects on the environment (Petron 1993, Martin and Coughtrey 1982, Ketchum et al. 1975).

Because of their relatively low population densities and frequently nocturnal habits, medium-sized mammals are difficult to study. Previously, the only technique to document movements of individual animals was radiotelemetry. We are evaluating a new application of radio frequency identification (RFID) technology. This technology allows us to remotely record the amount of time individual animals spend at a potential release site (PRS) for contaminants. The RFID system comprises a monitor, automatic data recorder, and remote camera system, which continuously monitor an unlimited number of different animal species at a specific site of interest. Once developed, this system can be used for studies of animal exposure to, uptake of, and transport of radioactive and nonradioactive contaminants anywhere animal exposure to point sources of contaminants is a concern, including LANL and other DOE facilities. Knowledge of contaminant levels and transport routes can be used to evaluate risks of contaminants to the environment and to examine options for environmental restoration.

RFID technology allows a very small (23 mm x 3.2 mm) glass-encapsulated "tag" to be implanted subcutaneously with a syringe and needle into animals. Tags last for the lifetime of the animal. As a marked animal walks through a portal monitor (figure 1), the monitor can record the identification number of the animal from the tag, along with the date



Figure 1. Picture of a squirrel just after it passed through the RFID portal monitor. The instruments mounted on posts in front of the monitor make and receive infrared beams. When an animal breaks a beam, it triggers a camera to take a picture.

and the time of passage. The technology is similar to a barcode reader. In combination with a remotely activated camera, this system records the time, date, and species as animals enter and leave a PRS and allows the investigator to determine the ratio of marked to unmarked animals of each species using a site.

The objectives of this project:

- Develop and evaluate a system that will allow LANL to efficiently document the amount of time spent by individual animals at a potential release site;
- Determine whether medium-sized mammals on LANL property exhibit elevated levels of metals or radionuclides; and
- Collect data during the evaluation process on the relationship between the amount of time an animal spends at a PRS and its level of contamination.

Method

The study was conducted at the radioactive liquid waste treatment lagoon for the Los Alamos Neutron Science Center (LANSCE) at Technical Area 53 (TA-53). LANSCE is a national user facility at LANL that provides intense sources of pulsed spallation neutrons for neutron research and applications. Operation of the linear accelerator and associated cooling systems produces radioactively contaminated liquid wastes. All potentially radioactive liquid wastes are collected and delivered to two sets of holding tanks. The waste is retained in the holding tanks until short half-life constituents decay completely. Tank contents are then pumped to the TA-53 radioactive liquid waste treatment lagoon. LANSCE is located on a mesa in the pinyon-juniper zone.

Between November 1996 and August 1997, medium-sized mammals (rock squirrels, raccoons, striped skunks, and grey foxes) were captured using cage traps. Animals were captured at TA-53 in two different areas (figure 2): within 400 m of the lagoon (at the lagoon), and more than 400 m from the lagoon (away from the lagoon). Animals were also captured in Guaje and Rendija canyons on Santa Fe National Forest lands. Captured animals were taken to an animal holding facility. There, we anesthetized them, inserted RFID tags under the skin between their shoulder blades, took measurements, and collected 2–4 g of hair. Animals were retained in the holding facility overnight and urine was collected in trays beneath the cages. We released animals at their capture site the next day. Urine samples were submitted to LANL's Health Physics Analysis Laboratory to be analyzed for radionuclide concentrations. Hair samples were submitted to the Laboratory's Inorganic Trace Analysis Group to test for concentrations of metals, including aluminum (Al), antimony (Sb), arsenic (As), barium (Ba), beryllium (Be), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), selenium (Se), silver (Ag) and thallium (Tl).

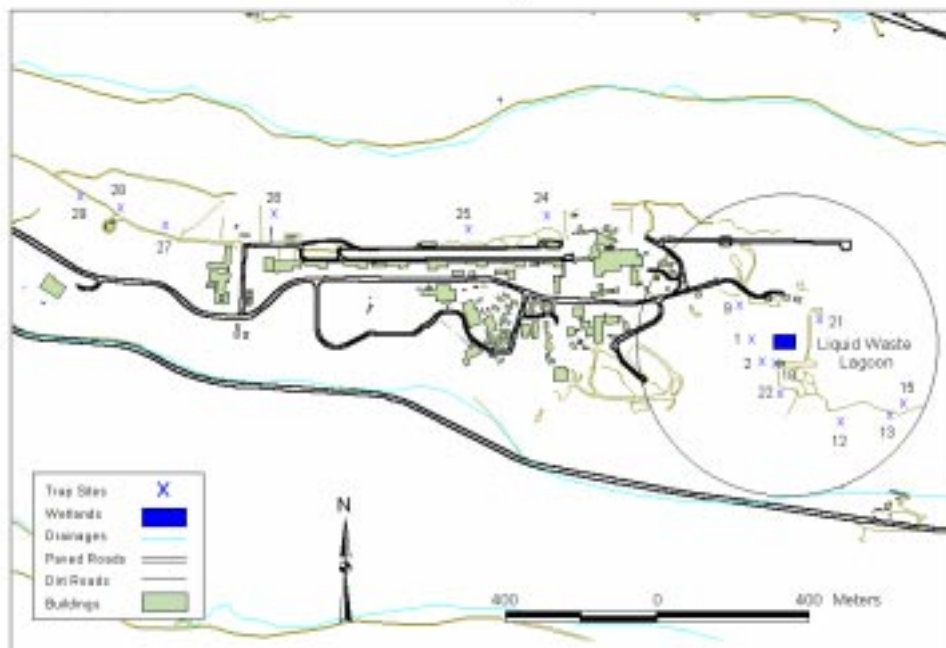


Figure 2. Locations of traps at TA-53. The circle is a 400-m buffer around the liquid waste lagoon. Animals captured inside the circle are considered to be at the lagoon. Animals captured outside the circle are considered to be away from the lagoon.

In April and May 1997, we used orange barrier fencing and chicken wire to construct an exclusion fence around the liquid waste lagoon. The RFID monitor was placed as an opening in the fence through which animals could access the lagoon. Trailmaster® cameras were placed to photograph any animal passing through the monitor. We operated the monitor and cameras continuously from May 15 through September 1 to document use of the lagoon area by marked and unmarked animals.

Progress and results

Animal Capture. Between November 19, 1996, and September 1, 1997, we captured 27 animals 58 times. Animals captured included 22 rock squirrels, two raccoons, one striped skunk, and two grey foxes. Twelve rock squirrels, two raccoons, and one striped skunk were implanted with RFID tags at TA-53 and released. Five rock squirrels were captured in Guaje and Rendija Canyons on Santa Fe National Forest land. Contaminant levels in these animals were used for comparison to levels in animals captured at TA-53. However, because of capture late in the fiscal year, one hair

sample from the control area was not submitted in FY97 and will be submitted in FY98.

Seven male rock squirrels were recaptured a total of 17 times, and four female rock squirrels were recaptured a total of 12 times. Male rock squirrels traveled an average of 185 meters between successive captures ($SD = 233$ m), with a minimum distance of 0 m and a maximum distance of 955 m. Female rock squirrels traveled an average of 110 m between successive captures ($SD = 98$ m), with a minimum distance of 0 m and a maximum distance of 281 m. There was no correlation between distance traveled and number of days between captures for males (Pearson's correlation coefficient; $r = -0.22$, $P = 0.39$, $n = 17$) or females (Pearson's correlation coefficient; $r = -0.13$, $P = 0.69$, $n = 12$). We did not detect an effect of the month of recapture on distance traveled (Analysis of Variance; $F = 1.71$, $P = 0.165$, $df = 6, 22$).

Effectiveness of the RFID monitor.

Of the 15 marked animals at TA-53, four animals (three rock squirrels, one raccoon) passed through the RFID monitor a total of nine times between May 15 and September 1, 1997. Two

pairs of readings appeared to represent rock squirrels entering and leaving the lagoon area. One animal remained in the lagoon area for 20 minutes; another remained for 2.5 minutes. We had five unpaired readings, indicating that either animals did not pass completely through the monitor or that they were able to enter or exit the lagoon area elsewhere.

We photographed seven rock squirrels, three raccoons, two cottontail rabbits, one woodrat, one chipmunk, one unidentified animal, and two birds at the monitor between May 15 and September 1. Of the nine passages through the monitor made by marked animals, only three were recorded by the camera system. Although some of these animals may not have passed completely through the monitor, we estimate that up to 50% of the smaller animals passing through the monitor were not detected by the camera. Of the two cameras used, one was operational for 94 days and one for 103 days out of a possible 111 days. We did not photograph any marked animals that were not recorded by the monitor, which suggests that the monitor approached 100% efficiency in detecting tagged animals.

Contaminants analyses. Urine was used in liquid scintillation and gamma spectroscopy analysis to measure the quantity of radionuclides. Detectable activity is defined as activity greater than two counting uncertainties of mean activity in blank control samples. None of the samples submitted for liquid scintillation analysis had detectable activity. Six of the samples submitted from 21 animals for gamma spectroscopy analysis had detectable activity: four animals at the lagoon, one animal away from the lagoon, and one animal from the control area. Three animals had ^{40}K in their urine (mCi/L ranging from 0.028 to 0.078) and one animal had ^{22}Na (0.011 mCi/L). The radionuclide present in the urine has not been identified for two animals (one animal away from the lagoon, one control animal). Two animals did not have sufficient urine for gamma spectroscopy analyses.

Median values of each metal in hair samples for each species captured on LANL are presented in table 1. We tested for significant differences in levels of metals in hair among locations (animals captured at the lagoon, animals captured away from the lagoon, and animals captured in the control area) for rock squirrels using Kruskal-Wallis tests (table 2).

Only aluminum, barium, copper, lead, arsenic, and selenium had a sufficient number of detections to test for differences in metal levels among groups. We examined differences among groups up to the $P < 0.15$ level to provide a very conservative test for differences. Levels of aluminum were significantly greater in the control animals than in animals captured at TA-53. Levels of barium may have been greater in the control animals also. Levels of selenium may have been greater in the animals captured away from the lagoon area at TA-53 than in animals from the control area or from the lagoon area. We did not detect significant differences in overall copper and lead levels among the three locations. However, there were a few individuals

captured at the lagoon area with high values of these metals (figure 3).

Conclusions and deliverables

The RFID monitor and tags were very effective at documenting animal passages into the lagoon area. The primary weakness in our design this year was our fence, which was penetrable. The low number of animals using the lagoon area surprised us. Only four marked animals out of 15 possible were documented as using the lagoon during May–August 1997. The absence of tritium in urine samples supported these results. We expected animals that used the lagoon frequently to be contaminated with tritium, but none of the animals we sampled had detectable contamination. We submitted two squirrels from the lagoon area for tissue analysis for tritium, which should provide more information on occurrence of tritium in these animals.

The radionuclide ^{40}K occurs naturally in soils around Los Alamos (Fresquez et al. 1997), and is not necessarily indicative of exposure to radioactivity in lagoon water. The presence of the radionuclide

Table 1. Number of animals with detectable values^a (n) and median values (μg/g) with range (in parentheses) of metals found in hair for animals captured at Los Alamos National Laboratory^b.

Metal	Rock squirrel (14) ^c		Raccoon (2)		Gray fox (2)		Striped skunk (1)	
	N	Median	n	Median	n	Median	n	Median
Al	14	90 (230)	2	345 (70)	2	325 (280)	1	690
Sb	2	0.35 (0.1)	2	0.8 (1.0)	0	N/A	0	N/A
As	5	0.2 (0.3)	1	0.2	1	0.2	1	0.3
Ba	14	8.1 (26.9)	2	8.75 (4.5)	2	4.65 (0.3)	1	15.0
Be	0	N/A	0	N/A	0	N/A	1	0.94
Cd	3	1.4 (0.3)	1	2.0	0	N/A	0	N/A
Cr	2	2.4 (2.0)	2	3.35 (0.5)	1	0.98	1	1.3
Cu	14	11.5 (52.9)	2	20.5 (15.0)	2	9.15 (1.5)	1	11.0
Pb	14	2.3 (25.3)	2	7.65 (7.9)	2	1.55 (0.9)	1	4.6
Ni	0	N/A	0	N/A	0	N/A	0	N/A
Se	13	0.7 (0.8)	2	1.95 (0.5)	2	0.8 (0)	1	1.2
Ag	1	2.3	0	N/A	0	N/A	0	N/A
Tl	0	N/A	0	N/A	0	N/A		N/A

^a Detection limits were: As, 0.2; Be, 0.07; Cd, 0.90; Cr, 0.9; Ni, 4.0; Se, 0.3; Ag, 2.0; Tl, 0.3.

^b All animals were captured at TA-53, except one fox that was captured at Area G.

^c The number of animals of each species tested (N) follows the name of the species in the table.

Table 2. Number of animals with detectable values^a (n) and median values (μg/g) with range (in parentheses) of metals found in hair of rock squirrels captured in three locations: at the lagoon, away from the lagoon, and at the control area.

Metals	At lagoon (11) ^b		Away from lagoon (3)		Control area (4)		Test statistic and P ^c
	N	Median	n	Median	N	Median	
Al*** ^d	11	190 (230)	3	170 (60)	4	835 (840)	8.8, P = 0.012
Sb	2	0.35 (0.1)	0	N/A	0	N/A	
As	5	0.2 (0.3)	0	N/A	4	0.2 (0)	6.0, P = 0.180
Ba*	11	7.7 (20.9)	3	15.0 (23.5)	4	21.5 (16.3)	4.4, P = 0.113
Be	0	N/A	0	N/A	4	0.12 (0.063)	
Cd	3	1.4 (0.3)	0	N/A	0	N/A	
Cr	2	2.4 (2.0)	0	N/A	1	1.2	
Cu	11	13.0 (52.7)	3	10.0 (2.9)	4	11.2 (6.4)	3.0, P = 0.223
Pb	11	4.9 (24.9)	3	1.6 (1.2)	4	2.4 (1.6)	2.4, P = 0.298
Ni	0	N/A	0	N/A	1	8.7	
Se*	10	0.65 (0.7)	3	0.9 (0.3)	2	0.7 (0.6)	3.8, P = 0.149
Ag	1	2.3	0	N/A	0	N/A	
Tl	0	N/A	0	N/A	0	N/A	

^aDetection limits were: As, 0.2; Be, 0.07; Cd, 0.90; Cr, 0.9; Ni, 4.0; Se, 0.3; Ag, 2.0; Tl, 0.3.

^bThe number of animals tested (N) in each location follows the location name in the table.

^cKruskal-Wallis test statistics and probability levels are included for those metals which had enough detections to test among groups.

^dMetals significantly different among groups at the P < 0.05 level were marked with three asterisks, at the P < 0.1 level with two asterisks, and at the P < 0.15 level with one asterisk.

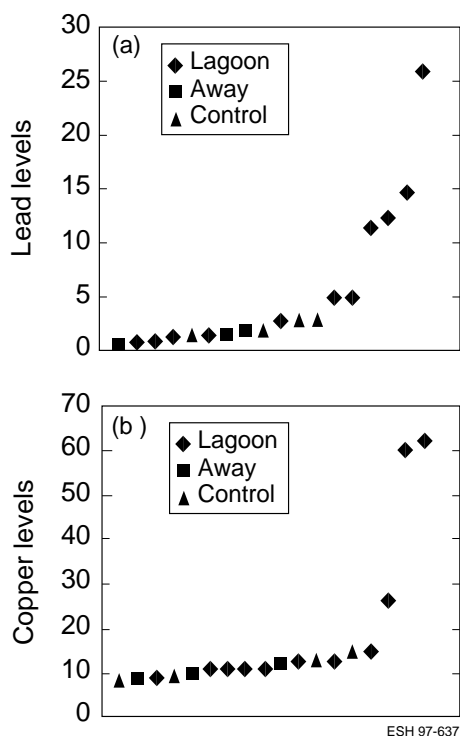


Figure 3. Levels of lead (a) and copper (b) in μg/g in hair samples from rock squirrels captured at TA-53.

²²Na probably does indicate uptake from the lagoon. Therefore, out of a total of 15 animals captured at the lagoon and potentially exposed to radioactivity from the lagoon, only one animal showed signs of radionuclide uptake from the lagoon water.

Toxicity from metals occurs in critical organs, frequently the liver, kidney, or brain. For most metals, there is often not a close correlation between metals in hair and metals in critical organs. Therefore, the analysis of hair samples for metal concentrations is primarily useful as a screening tool. We have little information on the significance of our aluminum, arsenic, barium, beryllium, or chromium values. Both aluminum and barium occur naturally at relatively high concentrations in soils of Los Alamos County (Ferenbaugh et al. 1990). Ranges of selenium observed in our study were

similar to those observed in a control population of raccoons (0.47–1.7 μg/g) used in a study in California, suggesting that our animals were not exposed to excessive levels of selenium (Clark et al. 1989). For animals in which cadmium was detected, concentrations in hair were as high or higher than in beavers in a industrially cadmium-contaminated river in Germany. No population-level effects were observed in the German beaver population (Nolet et al. 1994).

Although the median values of copper and lead in animals captured at the lagoon were not significantly different from those captured in the control area, we did have a few individuals with high values of these metals. The Kruskal-Wallis test is not sensitive to large outliers. Copper is a trace element needed by animals for proper physiological functioning (Robbins 1993). The highest copper levels we observed were similar to the highest recorded for domestic dogs in Japan (Hayashi et al. 1981). There is no indication that the levels of copper we observed indicate possible adverse effects. Lead in hair has frequently been used as an indicator of exposure. A safety limit of 9-μg/g lead in hair has been proposed for children (Revich 1994). Five animals (four rock squirrels, one raccoon) exceeded this level at TA-53 in our study; one rock squirrel had > 25-μg/g lead.

Liquid wastes contaminated with high levels of metals are not released into the lagoon. Therefore, any excessive metal concentrations observed in animals around TA-53 are the result of contaminants from historical activities and not from current use of the lagoon waters. We plan to continue this study for two more years and will collect additional data on contaminant levels under different environmental conditions.

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Seasonal Movements, Activity Patterns, and Radionuclide Concentrations of Radio-Collared Rocky Mountain Elk (*Cervus elaphus nelsoni*) Inhabiting the Los Alamos National Laboratory (LANL).

Principal Investigators: Phil Fresquez, James Biggs, Kathryn Bennett, Ecology Group (ESH-20)

Funding: FY96, \$68K; FY97, \$53K. The project also received \$28K from the Dual Axis Radiographic Hydrodynamics Testing facility mitigation action plan.

Introduction

According to estimates, over 1800 elk inhabit the Pajarito Plateau, including Los Alamos National Laboratory (LANL, Laboratory) lands, and this number appears to be increasing. Exactly how and where these animals use Laboratory property is unknown, but we do know that many of them use Laboratory technical areas. Some of these technical areas contain environmental contaminants that may ultimately constitute a pathway by which radionuclides can be transported to people (Fresquez et al. 1994).

Several neighboring pueblos—Santa Clara, San Ildefonso, Cochiti, and Jemez—have voiced concern to the Laboratory Director about this potential environmental problem. Additionally, public concern over increased elk use and potential human-animal conflicts has resulted in the formation of a regional “elk study group” involving representatives from LANL, the Department of Energy (DOE) Los Alamos Area Office, DOE Albuquerque, the US Forest Service, the US Park Service, Los Alamos County, and the New Mexico Department of Game and Fish. At the request of a US state representative, this committee was formed to address human safety issues with elk.

Our study supports the Laboratory by helping it meet DOE Orders 5400.1 and 5400.5, the Resource Conservation and Recovery Act, the Clean Water Act, the New Mexico Water Quality Act, and the National Environmental Policy Act. The information collected in this study will enable in-depth assessments of large-game mammal ecology for use in a sitewide Environmental Impact Statement (EIS), project-specific EISs, and Environ-

mental Assessments. This information may also aid the Laboratory in addressing concerns raised by the public during September 1997 scoping meetings. Furthermore, a series of interagency meetings between LANL, the state, and the US Park Service has identified the need for data on this species to develop a regional management plan.

To accomplish an accurate radiological study, more information is needed about the numbers of resident and migratory elk on Laboratory property, the locations of elk on these lands, and most importantly, radionuclides in tissues of animals that have a **defined history of using** (i.e., the area and amount of time an elk spends in an area) LANL technical areas. A management plan is necessary to address these issues; however, data on habitat use, activity patterns, and resource use of elk on LANL properties are inadequate to develop an appropriate management plan that addresses current and anticipated future problems.

The FY96 and FY97 objectives of this study are

- evaluate global positioning system (GPS) test collar data in various plant communities and terrain;
- apply spatial and temporal analysis of data to evaluate activity patterns of elk;
- compare radionuclide content in collared elk from LANL to background locations;
- calculate the committed effective dose equivalent and the risk of excess cancer fatalities to people who consume meat from elk that use LANL lands;
- develop habitat use models for use in the National Environmental Policy Act process and to develop long-term management strategies; and

- identify locations of concern for development of management recommendations to aid local agency officials in minimizing human/animal conflicts.

Before we initiated this study, the most common method for collecting data on wild animal activity was by tracking them with very high frequency (VHF) radio collars. Although this method is still the most widespread, new methods, including the use of GPS technology, are being tested. We elected to try GPS collars due to inherent problems with conducting a study of this magnitude using VHF. Among these problems are inaccessibility of secured areas on a regular basis; inaccessibility of areas due to terrain; land ownership and political boundaries; the high-cost requirement of human resources; and inaccuracies of data associated with triangulating positions (the act of obtaining a locational fix by taking a ground reading from at least two points, preferably three).

When we initiated our study, GPS collars included several types. We were among the first to test and use a model developed by Telonics, Inc. (Mesa, Arizona), a leader in the wildlife telemetry technology field. Because these collars are self-activated and store and disseminate data from the collar, personnel are not needed in the field to collect data. Consequently, time and cost savings have been significant as shown in table 1.

To date and with the technology available in GPS collars as compared with that of VHF systems, we would realize savings of approximately \$150K. As the number of animals collared increases, so do cost savings.

Table 1. GPS/VHF Collar Cost Comparison.

	GPS Collar (n=6)	VHF Collar (n=6)
Person Costs (1 fix/day for 6 animals)	\$40/day (data downloading)	\$560/day (avg. of 1 fix/day)
Total Person Cost for 12 Months of Tracking	\$14,000	~\$200,000
Collar Cost (6)	\$32,000	\$2,100
Receiver Cost	\$1,750	\$500
Data Analysis	\$14,250	\$15,000
Other Cost	\$6,000	\$250
TOTAL COST	\$68,000	\$217,850

Method

To achieve the objectives of this study, we are employing two methods to obtain data:

- GPS radio collar data/Geographical Information System (GIS) interfacing in the application of radio telemetry for continued tracking of elk movements and evaluation of resource use on LANL and adjacent property and
- Tissue samples collected for radiological contamination studies.

Trapping. Three sites in two areas of LANL were selected for trapping elk based on three criteria: 1) Previous movement data of elk on the pajarito Plateau: We attempted to collar animals of different herds at different locations on LANL property to maximize representation of elk populations in this area; 2) Known areas of high elk activity: Due to labor, time, and budget constraints, we had to maximize our probability of capturing animals within a given time period; and 3) Proximity of trapping to areas of current LANL operations/resource conflict issues: Although not reported in this paper, a secondary objective of this study was to identify potential pathways of contaminant transport off LANL property; therefore, trapping locations were located near a radioactive-waste burial site and outfall effluent sources (artificial water sources).

We have deployed GPS collars on seven elk to date. We have collected data on physical measurements, blood diseases, tritium concentrations in blood, age, weight, and reproductive condition of each animal. Trapping took place from January through April 1996. This time period maximized the chances for capture because natural food sources were less available due to snow cover and it was before the beginning of seasonal migration. Animals were captured with clover traps baited with apples and alfalfa. Animals were restrained by lowering the animal to the ground within the clover trap using ropes. Once the animal was restrained, trained personnel entered the trap and placed a hood over the head of the animal. Animals were then fitted with the radio collar.

GIS coverages/GPS locational position overlays. Habitat use and availability are being evaluated by overlaying elk locations on a vegetation land cover map that delineates dominant overstory vegetation. Ground-truthing LANDSAT thematic mapper images that detect reflected radiation from the earth's surface (infrared wavelengths) were classified into five land-cover types used for this study: unvegetated/developed lands; mixed-conifer forests; ponderosa pine forests; pinon-juniper woodlands; and grass/shrublands.

Results

Test collar. As part of the initial stages of this study, we programmed one collar to collect locational data every 20 minutes to evaluate its usefulness under various plant canopy closures and terrain. Following differential correction at each of the test sites, we determined the locational error of the GPS collar by making comparisons between the test collar and a hand-held GPS. The overall mean locational error of the GPS collar was 106 ± 16 m (figure 1); therefore, we applied this error rate to the collars deployed on the elk. Furthermore, there were no statistically significant differences ($\alpha = 0.05$) in locational errors between ponderosa pine and piñon pine-juniper vegetation types ($p = 0.8199$); open and closed plant canopies ($p = 0.8672$); or canyons and mesa tops ($p = 0.9874$). We also found no significant differences in the observation rate, i.e., the number of positions obtained in a defined period of time divided by the maximum number of positions possible, of the collar in each of the plant cover and terrain types. Although we found no differences in the collar's effectiveness under different environmental conditions, we are planning to deploy additional test collars under similar conditions to further test the collars effectiveness.

Elk/habitat use. We programmed all elk-deployed GPS collars to locate positions every 23 hours. Between April 1, 1996, and August 30, 1997, we collected >1900 fixes with an approximately 75% observation rate (range 60–90%). We successfully interfaced GPS fixes of elk locations and detailed vegetation maps using the GIS to provide seasonal data (spring, calving, summer, fall, winter) on habitat use within and immediately adjacent to LANL (figure 2). Based on habitat use and availability analysis, use of grass/shrub and pinon/juniper habitats was generally higher than expected during most seasons and use of forested habitats (ponderosa pine, mixed conifer) was lower than expected.

Elk habitat use/movement predictive model. As part of the overall study, we will prepare a model that predicts alterations in habitat use and movement

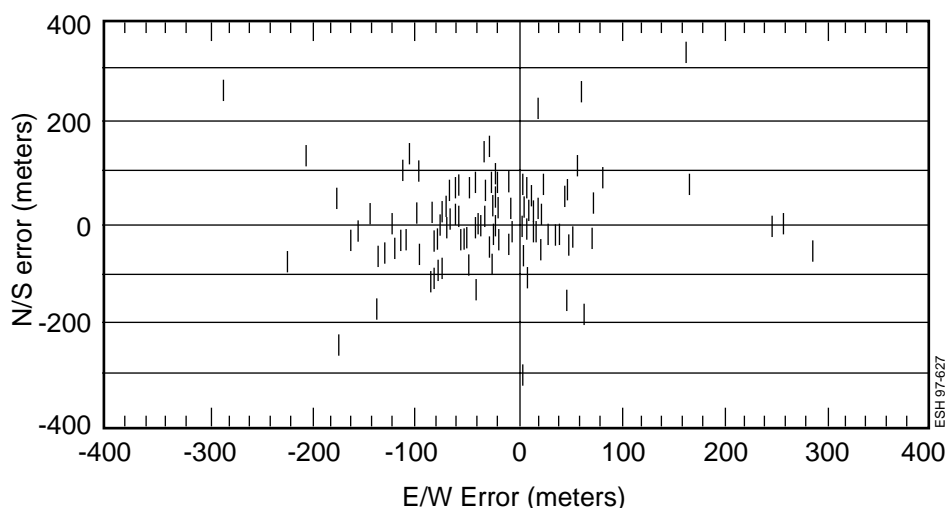


Figure 1. East-west and north-south error calculated from the collar locations

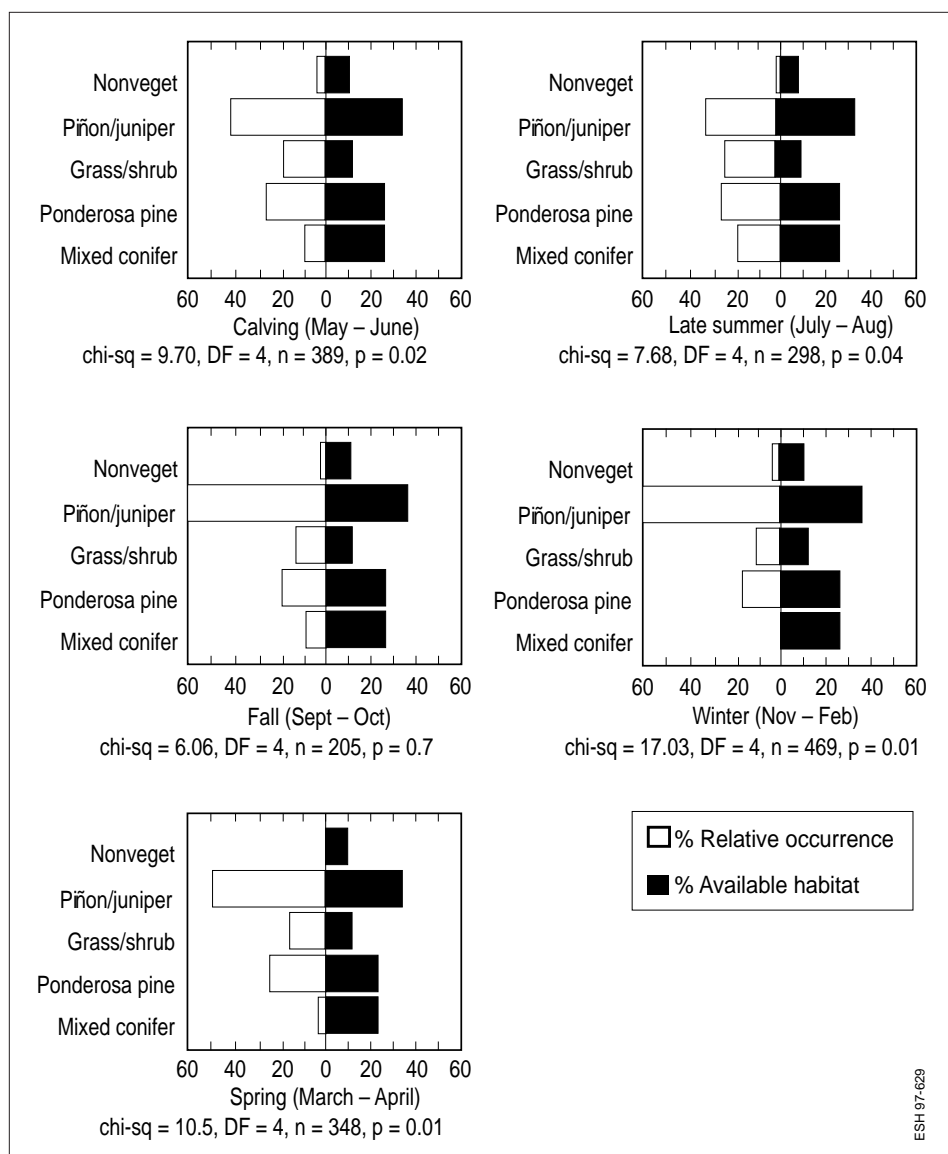


Figure 2. Habitat use and availability in and around LANL

patterns due to changes in the environmental landscape, either as a result of natural disturbances, i.e., fire, or human-induced disturbances, i.e., fences, buildings, roads. We have initiated development of the model by interfacing GPS locational positions of elk with GIS-produced overlay coverages. A least-cost surface analysis model was used to identify elk pathways. This model was developed in the ARCVIEW extension Spatial Analyst by comparing locational positions to slope, aspect, vegetation cover type, proximity (including proximity to water sources), type, and influence upon developed areas.

We are still evaluating additional variables and have identified new variables that we have only recently suspected as being potential influences based on our recent data analysis. These variables include secondary and tertiary roads and associated traffic patterns, the construction of facilities/projects in potential high-use or specialized-use habitats (watering, calving, bedding), the use of disturbed vs. undisturbed habitats (including potential contaminant release sites), and the effects of external “pressure” locations, i.e., hunting at San Ildefonso Pueblo, on distribution patterns of elk on LANL property. When we input the identification, evaluation, and application of these variables to the model, it will make its predictive power much more accurate. Furthermore, during the process of evaluating these variables, adjoining land management organizations, such as Native American Pueblos and Bandelier National Monument, can use the information during the development of species management plans.

The greatest benefit of the model development and its subsequent testing and refining will be that a tool will be available that will aid in identifying potential hazards associated with altering activity patterns of elk on LANL property from LANL projects. Also, during the testing and refinement of the model, specific activity pattern and habitat use data will be identified and tested that can be used in the management plan of elk, which is viewed as a future necessity for LANL. A conceptual design of the model

depicting input variables is shown in the diagram to the right.

The model threshold distance has been defined not to exceed the seasonal daily distance traveled by elk. The model also evaluates the requirement of water sources in the path selection.

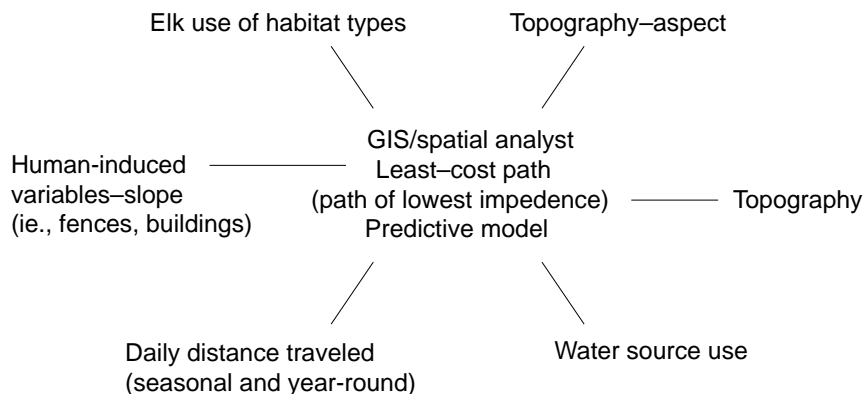
Elk/home range analysis. We have been examining estimated yearly and seasonal home ranges of the GPS-collared elk that appeared on Laboratory property at least partially between February 1996 and September 1997. The estimates will help us determine areas of concentrated use and significant factors, such as terrain and habitat, in the distribution of elk. Seasonal home ranges (95% utilization distributions) varied from 903–5004 ha during winter, 1218–6157 ha in spring, 2138–7907 ha during calving, 1957–3306 ha in summer, and 3021–10,160 ha in fall (Figure 3). We found that seasonal core activity areas (50% utilization distributions) were generally less than 500 ha for most animals.

We are also using the evaluation of home-range size to look at the importance that habitat type and terrain use has on this species during various times of the year, i.e., calving, wintering, which will help in the development of an overall management plan. Additionally, the analysis of the data will also help us identify biases associated with observation rates of GPS collars by comparing the known positional fixes to missing data points within the home range.

Elk/water resource use. Due to the Laboratory initiative to reduce the number of effluent outfalls, we have been evaluating relative percent seasonal and yearly water source use by elk to identify the role this resource plays in every day elk movement patterns throughout the Laboratory. We have been analyzing water use by overlaying perennial water sources onto the GIS and calculating the number of elk locational positions within a set of five distances from that source: 0.25 mi, 0.50 mi, 0.75 mi, 1.0 mi, and 2.0 miles. Based on analysis of two perennial water sources on LANL property, we have found that cumulative percent use

The model equation is defined as follows:

$LCP \text{ (least cost path)} = HT \text{ (hab. type)} + Asp \text{ (aspect)} + Slp \text{ (slope)} + Hvar \text{ (human vars.)}$.



was 17%, 35%, 49%, 61%, and 90%, respectively. We will continue to identify elk use around other water sources on LANL property.

Tissue/radiological concentrations.

As part of the scope of work outlined in the FY96 and FY97 proposals, we proposed to track GPS-collared animals

for approximately one year. At the end of this time, the plan was that the animals would be sacrificed and their meat and bone collected for the analysis of various radionuclides (^3H , ^{241}Am , total U, ^{137}Cs , ^{90}Sr , ^{238}Pu , and ^{239}Pu). Since the onset of the study, we have retrieved samples from three of four killed animals: two

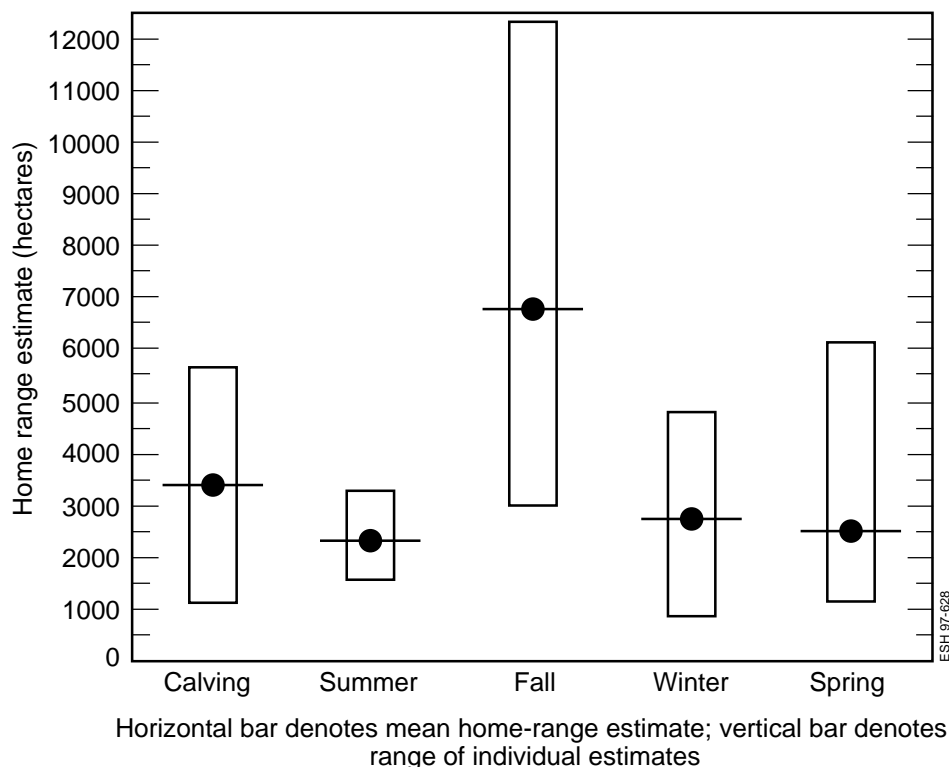


Figure 3. Home range estimates of six GPS-collared elk on or near LANL, February 1996–September 1997

from hunters and the third (bone sample only) from a roadkill. We were unable to retrieve samples from one of the harvested elk.

We have not yet received most of the analyses back from the chemistry laboratories. Of the data that have been received, the radionuclide concentrations in muscle and bone show that a few radionuclide concentrations, particularly in bone samples, are just above background levels; however, this does not pose any hazards to people who consume the bone and especially the meat. We expect the rest of the analyses on these animals by this winter. Once we obtain the analyses, we will trace the history of the elk's movement on LANL property during the course of the study period.

We have an additional two animals still alive that have moved on or been located immediately adjacent to LANL property. We are in the process of harvesting these animals. We will also look at the history of their movement on LANL property. These data will be imperative to giving conclusive findings about animal movement on LANL property and the associated contaminant levels.

Conclusions and deliverables

GISs have developed as an important research and information tool and, combined with GPS location of animals, satellite data can be easily and quickly incorporated into graphics, such as detailed maps. Additionally, data can be statistically analyzed and modeled. The recent development of GPS units on radio collars has allowed us to obtain more data at a lower cost. Using the innovative GPS radio collars, we've been able to do more intensive tracking, obtain more frequent and accurate locational data, and will be better able to model activity patterns. Because GPS represents a new technology, the evaluation of its usefulness and application are at the forefront of research on wildlife telemetry systems. Many future applications of GPS technology to wildlife studies will be based on work being conducted currently by a small number of researchers, including ourselves.

The GPS collar has just recently been put on the market for use in studying wildlife species; consequently, few studies have been conducted that examined the collar's effectiveness in various environments. Until TDEA funded this study in the mountainous terrain of Los Alamos county, no previous studies examined the collar's use in the western US. Through our research, we have determined error rates of GPS collar performance according to varying vegetation and terrain types. These rates can be applied to locational fixes of animals when the collars are deployed in an applied field study. The results of our research exceed early expectations of the collar's performance potential and have generated a tremendous amount of interest among natural resource management organizations.

Furthermore, we have been able to identify specific activity patterns of elk on the Pajarito Plateau with much greater accuracy and precision than would have been possible with other forms of tracking devices. Our studies have identified habitat use patterns, water source use patterns, home ranges, and movements based on human-induced barriers. We have also successfully interfaced the GPS data with GIS to further advance the applications of the technology.

The testing and successful application of a new form of technology, such as the GPS collar, have already begun to benefit the collection of wildlife data within the boundaries of LANL. Our current work has received much attention in the wildlife management and research community. To date our work has provided several state natural resource agencies, universities, and international wildlife monitoring departments enough preliminary data for them to determine if this GPS technology would prove useful for their endeavors. Several organizations have contacted us to evaluate the potential for collaborative studies on the use and application of GPS for studying other wildlife species.

Two earlier technical evaluations of the GPS collar appear in previous reports:

- the testing of a GPS-borne radio collar for wildlife use (Bennett et al. 1996), and
- the deployment and application of a GPS collar to wildlife studies (Biggs et al. 1996).

In addition, we have made several technical achievements during the course of this project.

We have set the following objectives for our research in FY98:

- further develop the predictive model based on recently identified habitat and human-induced variables;
- test and verify the model via additional collar deployment, data acquisition, and habitat variable measurements;
- develop risk analysis of humans who consume elk meat based on tissue analysis of samples collected from animals used in our study; and
- increase sample size to establish a baseline of resource use that will be included in developing initial management strategies for elk on or near LANL property.

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Health Physics



*Applications of Thermal Ionization
Mass Spectrometry (TIMS) to the
Detection of ^{239}Pu and ^{240}Pu Intakes*

*High-Energy Neutron Dosimetry and
Spectroscopy*

*Development and Implementation of
the Los Alamos National
Laboratory (LANL) Neutron Extremity
Dosimeter*

*Optimization of Continuous Air-
Monitoring (CAM) Instrument
Placement*

Resuspension of ^{238}Pu from Surfaces

Applications of Thermal Ionization Mass Spectrometry (TIMS) to the Detection of ^{239}Pu and ^{240}Pu Intakes

Principal Investigators: W. C. Inkret (PI), G. Miller (Col), D. Lewis, Radiological Dose Assessment (ESH-12); D. W. Efurd (Col), M. Attrep, Jr., T. M. Benjamin, D. E. Dry, C. A. Hensley, D. L. Kottman, F. R. Roensch, Nuclear and Radiochemistry (CST-11); D. J. Rokop (Col), H. Poths, Environmental Systems and Waste Characterization (CST-7); D. L. Wannigman Operational Health Physics (ESH-1)

Funding: FY96, \$61K; FY97, \$61K

Introduction

Current US Department of Energy (DOE) regulations require routine bioassay monitoring for all workers who have a reasonable potential for intakes of radioactive materials that may result in a committed effective dose equivalent (CEDE) of 1 mSv in a single year. The concepts of committed dose equivalent and CEDE are discussed in National Council on Radiation Protection and Measurements (NCRP) Report No. 84 (NCRP84). The NCRP considers these quantities useful for radiation protection planning and for demonstrating compliance and recommends their use for such purposes. The concept of committed dose involves the extrapolation of cumulative dose to 50 years into the future. The term "monitoring" is defined in the Code of Federal Regulations (10CFR835) as "the measurement of quantities...of radioactive material and the use of the results...to evaluate potential and actual exposures."

During FY96, the project discussed in this report established a viable urine bioassay program using TIMS for detecting occupational intakes of ^{239}Pu and ^{240}Pu . TIMS analysis was put into routine use on January 1, 1997.

The purpose of this year's project, analysis on the two-stage TIMS instrument, is to develop an even more sensitive plutonium detection capability by removing noise components found in the single-stage TIMS process.

Method

Urine *excreta* were collected for a 24-hour interval from a population of individuals who work at Los Alamos National Laboratory (LANL). Each

24-hour urine sample was traced with ultrapure ^{242}Pu (Johnson 1977). The plutonium fraction was coprecipitated with alkaline earth phosphate at room temperature. The precipitate was dissolved in 8-M HNO_3 , heated, and adsorbed onto anion exchange resin. The plutonium was eluted from the exchange column with successive rinses of 0.5-M HCl , HI-HCl reagent and H_2O . The elute was then evaporated to dryness. The plutonium was dissolved in 12-M HCl containing a drop of H_2O_2 and adsorbed onto a second anion exchange column. The column was rinsed with the HI-HCl reagent. The plutonium was then electroplated onto a platinum disk. The platinum disk was analyzed under vacuum with a 300-mm² solid-state surface barrier detector for 10,000 minutes. The plutonium was removed from the platinum disk with HF and HNO_3 . The sample was evaporated to dryness. The plutonium was dissolved in 12-M HCl containing a drop of H_2O_2 and adsorbed onto a third anion-exchange column. The sample was rinsed with 8-M HNO_3 and the plutonium was eluted with a rinse of HB_r . The sample was deposited on a platinum wire filament and analyzed for ^{239}Pu and ^{240}Pu atom contents by TIMS. All chemical processing was performed in class-100 clean rooms (Moody 1982).

Chemical and TIMS analyses were performed on six blank synthetic urine samples to determine the best background subtraction protocol for low-level samples. Two protocols were evaluated: continuous background data collection during sample analysis; and discrete collection at the beginning and end of sample analysis. A set of 12 human urine

samples were collected, processed for introduction into the two-stage spectrometer, and analyzed on the two-stage TIMS. The urine samples were collected from individuals with little or no-known occupational exposure to plutonium.

Progress and results

Synthetic urine. Chemistry and TIMS analyses were performed on six blank synthetic urine samples to determine the best background subtraction protocol for low-level samples. Two protocols were evaluated: continuous background data collection during sample analysis and discrete collection at the beginning and end of sample analysis. Results indicated a lower uncertainty associated with the continuous collection protocol.

The synthetic urine samples showed the presence of isobaric interferences at about the same magnitude as noted in human urine samples. However, the 3.7 mBq per 24-hour bias noted in the samples set listed in table 1 and displayed in figure 1 was not observed in the synthetic urine. One interpretation of this result is the presence of environmental levels of plutonium in the human samples.

The presence of the isobaric interferences appears to be unrelated to the specific urine matrix of human versus synthetic urine. This result indicates that variations in isobaric effects may be due to the quality of reagents and ion exchange resins used in chemical preparation of urine samples. These results also indicate that the resolution of the two-stage TIMS should have sufficient sensitivity to resolve the isobaric hydrogen mass defect.

US DOE Office of International Health Studies (OHIS) intercomparison study. Participation in the DOE Office of International Health interlaboratory comparison for detecting ultralow levels of plutonium in human urine was completed. In general, the results were promising and certainly validated the use of the single-stage TIMS instrument for monitoring plutonium workers. The synthetic urine samples were chemically prepared with the same procedure used for human urine samples. The prepared synthetic urine samples were analyzed with alpha-spectroscopy and TIMS. No activity was detected by the alpha-spectroscopy analysis for any of the synthetic urine samples. Figure 2 contains a plot of the TIMS measured plutonium activity in each sample as a function of the known activity. Detailed results are available in a NIST report (NIST 1997).

Note the increase in the relative measurement uncertainty with increasing measured activity in figure 2. Figure 3 contains a plot of the uncertainty of the TIMS measured ^{239}Pu activity as a function of the true value of ^{239}Pu activity. The estimated measurement uncertainty for the blank synthetic urine samples was $4.8 \text{ mBq} \pm 0.6 \text{ mBq}$. This uncertainty corresponds to a classical minimum detectable activity (MDA) of 22 mBq and an estimated minimum detectable CEDE of 2.5 mSv.

Human urine and ^{242}Pu tracer. The ^{242}Pu tracer used in the single-stage TIMS process has been analyzed on the two-stage instrument. The results indicated that the ^{242}Pu tracer in stock at LANL is a suitable tracer at levels that are lower than achievable on the single-stage TIMS instrument. This initial review indicated that the sensitivity for bioassay analysis would not be limited by the isotopic purity of the ^{242}Pu spike.

Direct loading of the ^{242}Pu on the two-stage TIMS obtained a $^{242}\text{Pu}/^{239}\text{Pu}$ atom ratio of 3.2×10^8 and a $^{240}\text{Pu}/^{242}\text{Pu}$ atom ratio of 5×10^8 . It appears that the ^{242}Pu tracer has more ^{240}Pu than ^{239}Pu impurity than was expected from the initial measurements on the tracer material. This

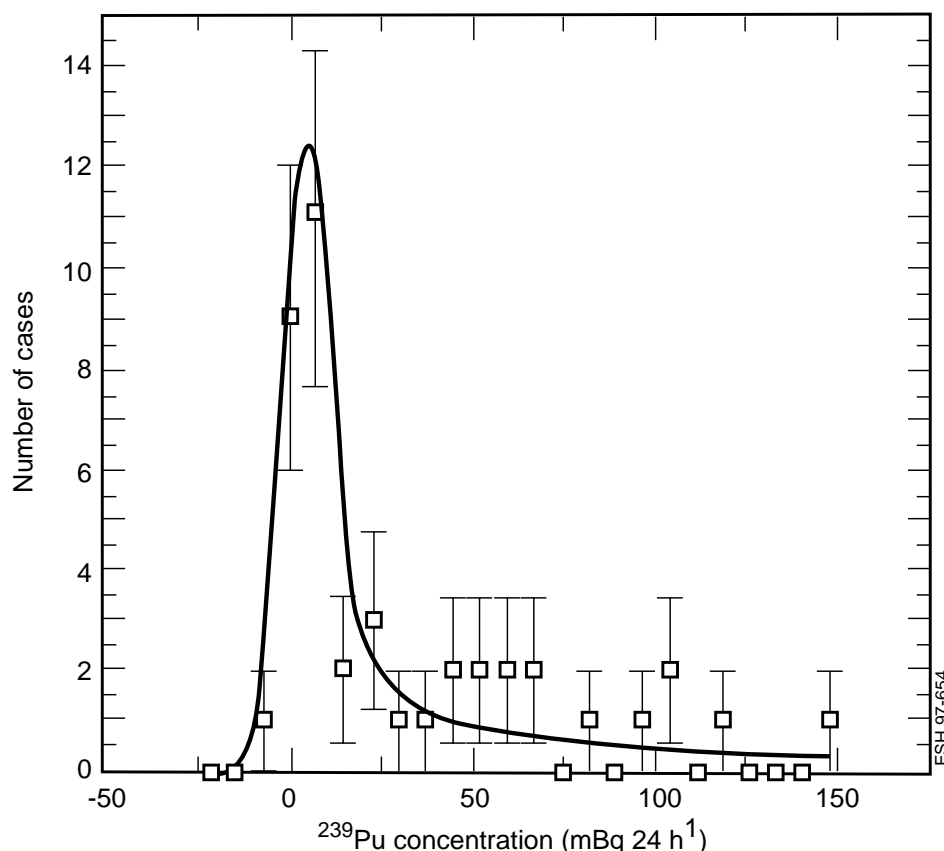


Figure 1. Histogram of TIMS ^{239}Pu urine bioassay measurements from a population of workers at LANL. The solid line represents a convolution of a Gaussian measurement error distribution and a delta-uniform prior distribution, (the uniform is in log space). The error bars represent the binned data uncertainty as the square root of the number of events in a bin. The TIMS measurement error based on the fit is $3.81 \text{ mBq} 24 \text{ h}^{-1}$.

result gives a theoretical classical MDA lower bound of 1.6×10^5 atoms of ^{239}Pu using a 2-ng spiking of the sample. We did not have the resources to determine the effect of using smaller quantities of ^{242}Pu tracer. A simple arithmetic average of the single-sample uncertainties yielded a theoretical limit of the classical MDA of 1.9×10^6 atoms of ^{239}Pu and a theoretical limit of 2.7×10^6 for ^{240}Pu .

Statistical evaluation of TIMS data. Table 1 contains measured ^{239}Pu concentrations for the sample group of individuals at LANL. Individuals from this group who have worked with plutonium are indicated in table 1. Table 1 also contains the collected and analyzed urine volumes. The average volume of urine collected over a 24-hour period was 1800 mL and the population standard deviation was 800 mL.

Figure 1 contains a histogram of the measurement results. Figure 1 also contains a nonlinear fitted plot of a convolution of a Gaussian measurement error distribution and a two-component delta function-uniform prior distribution (uniform in log space) (National Council on Radiation Protection c14). The delta function represents the nonoccupationally exposed population and the uniform distribution represents the occupationally exposed population (Miller 1993, 1995). The estimated measurement uncertainty through the ultratrace chemistry and the TIMS instrument process is 3.8 mBq per 24 hours. This result compares well with the average single sample uncertainty for the data points 1 through 58b. Table 2 contains a comparison of the LANL TIMS and alpha-spectroscopy measurement capabilities for ^{239}Pu . The distributional analysis revealed an

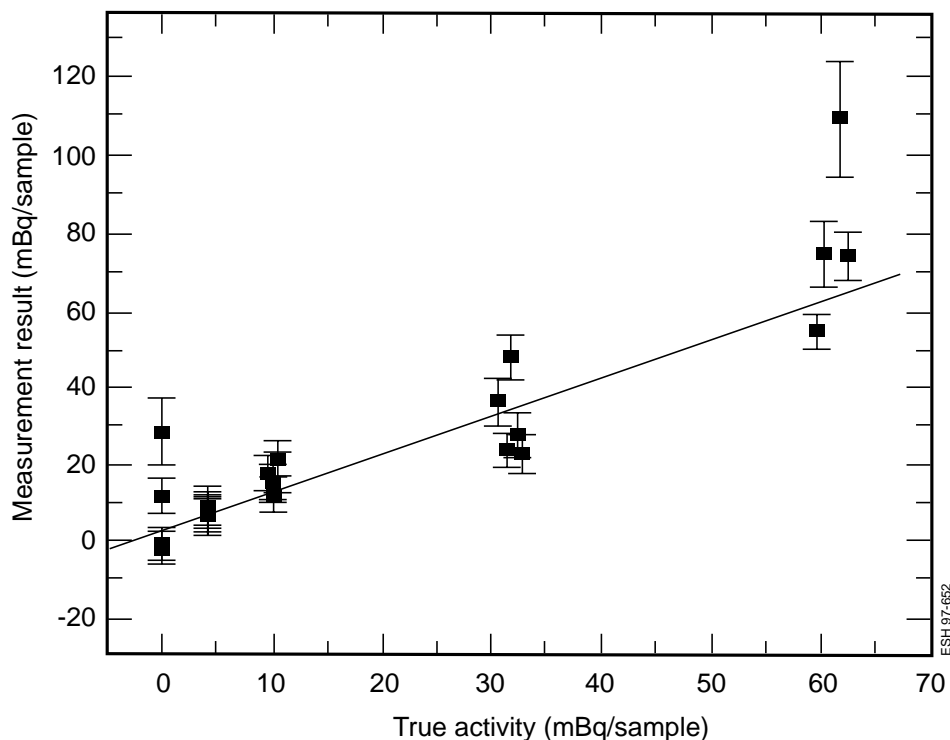


Figure 2. Measured ^{239}Pu activity as a function of true sample activity. Results are from the TIMS analysis of synthetic urine samples. The line indicates a simple linear-weighted fit of the data. The linear fit has a intercept of $3.3 \mu\text{Bq} \pm 2.2 \mu\text{Bq}$, a slope of 0.98 ± 0.09 and a correlation coefficient of 0.92.

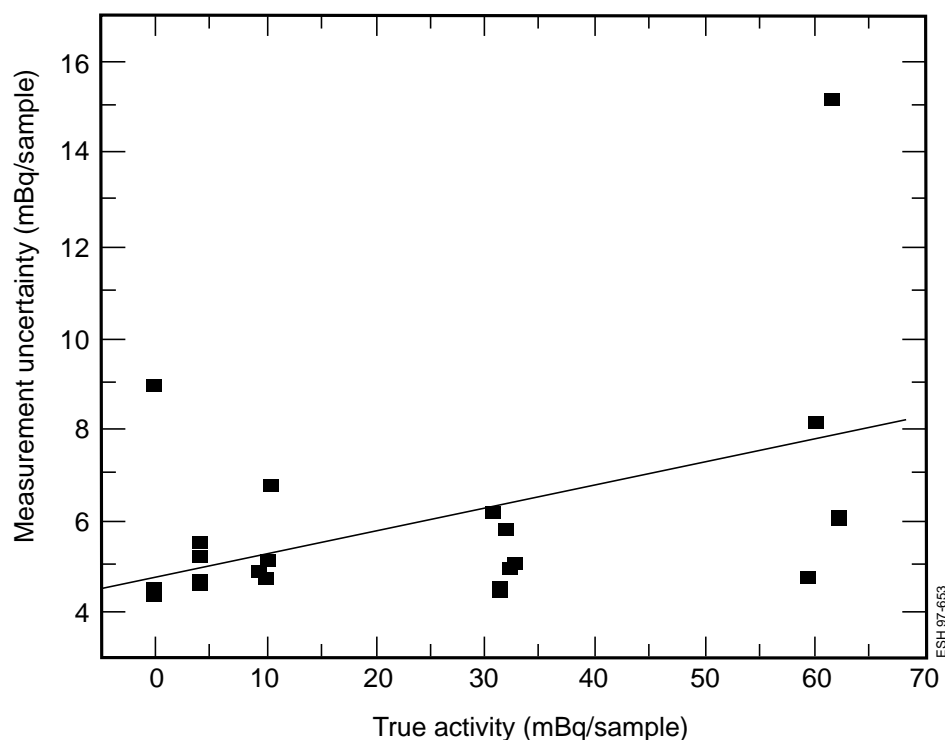


Figure 3. Uncertainty of TIMS-measured ^{239}Pu activity as a function of true sample activity. The line indicates a simple linear fit of the data. The linear fit has a intercept of $4.8 \mu\text{Bq} \pm 0.06 \mu\text{Bq}$, a slope of 0.05 ± 0.02 and a correlation coefficient of 0.48.

estimated measurement bias of 3.7 mBq per 24 hours for the ultratrace chemistry and TIMS measurement process.

Instrumentation electronics. Results from the synthetic urine and blank studies indicate that diurnal changes were occurring in the dark current of the mass spectrometer electronics. One of the major sources of this dark current is due to the differential pumping system associated with the detectors. Early morning pressures were typically 2×10^{-9} torr and a dark current signal of approximately 15 to 20 counts per 100-second count interval. Afternoon pressures were typically 6×10^{-9} torr and a dark current signal on the order of 50 counts per 100-second count interval.

The electronics system for the detector portion of the mass spectrometer was reevaluated and upgraded. The upgrade included evaluating the electron multiplier for poor electronic connections that could cause spurious counts to be recorded. The final result was a more uniform dark current throughout the day and a dark current count rate of about 25 counts per 100-second count interval.

The reduction of dark current improved the precision of routine TIMS sample analyses and has allowed a shift of focus to the problem of isobaric interferences. The improvement in the precision from this project has benefited both the LANL occupational worker bioassay monitoring program, the environmental monitoring program, and other technical programs that use the TIMS measurement technology.

Discussion and conclusions

The radiochemical alpha-spectroscopy technique used at LANL for plutonium urine bioassay has a measurement uncertainty of 150 mBq per 24-hour urine sample (Miller 1995). This measurement capability corresponds to detecting an intake of 1 kBq, and a resulting minimum detectable CEDE of 90 mSv, if the measurement is made at 180 days after an acute inhalation intake of 1- μm AMAD, inhalation class Y, ^{239}Pu .

Table 1. Summary of ^{239}Pu urine assay results for a population of LANL workers. The urine samples were processed in class-100 clean rooms using ultratrace actinide chemistry procedures, and analyzed on a thermal ionization mass spectrometry instrument. Results from this table are displayed in the histogram in Figure 1. In cases where two samples were submitted by one individual, the samples are denoted by the letters A and B. Urine samples were collected over a 24-hour period. Individuals who have worked with plutonium are indicated in the last column by the letter Y.

ID	^{239}Pu urine concentration (μBq24h^{-1})	σ (μBq24h^{-1})	Analyzed volume	Collected volume	Plutonium work?
1	-0.7	0.4	1750	1990	Y
2	512.8	7.0	995	995	Y
3	67.0	44.4	1500	1835	Y
5	35.6	4.1	1865	1865	Y
9	-0.5	2.6	2395	2395	Y
10	-8.4	5.2	1750	3030	Y
14	3.0	3.0	1845	1845	Y
15	36.4	7.0	1500	3270	Y
17	9.1	3.0	1620	1620	Y
20	95.0	5.2	1750	2735	Y
21	62.0	3.7	1750	1995	Y
22	25.5	4.1	1730	1730	Y
23	47.4	5.2	1750	1985	Y
24	5.8	3.0	1530	1530	Y
26	6.8	3.0	535	535	Y
27	12.4	3.7	1142	1142	Y
31	-1.2	4.4	1750	2720	Y
32	105.8	3.3	680	680	Y
36	3.7	3.0	1065	1065	Y
37	79.0	4.8	1750	2590	Y
38	146.4	4.8	1070	1070	Y
40	1.6	4.1	1750	2135	Y
41	5.9	5.2	1500	2860	Y
42A	8.0	3.3	1340	1340	Y
42B	4.8	3.0	355	355	Y
43	19.2	5.6	1450	2920	Y
44	0.2	4.4	1750	2430	Y
45	115.1	5.6	1210	1210	Y
46	7.8	3.0	672	672	Y
47	439.9	7.0	1515	1515	Y
49	105.4	13.3	1710	1710	Y
51	47.3	2.6	2465	2465	Y
52	50.8	2.2	2358	2358	Y
53	48.4	3.7	925	925	Y
54	39.1	3.0	3120	3120	Y
56	6.6	2.2	1258	1258	Y
57	13.6	2.2	3155	3155	Y
58A	-0.6	2.2	987	987	Y
58B	10.3	2.6	910	910	Y

Application of the class-100 clean room radiochemistry and the single-stage TIMS to the determination of plutonium concentration in the collected urine samples yielded an estimated measurement uncertainty of 3.8 mBq per 24 hours. The estimated bias for the TIMS analysis is 3.3 mBq per 24 hours. The current TIMS measurement capability for human urine corresponds to detecting an acute inhalation of 30 Bq of ^{239}Pu for annual routine monitoring and a resulting minimum detectable CEDE of 2 mSv.

The result for human urine compares favorably with the estimated synthetic urine measurement uncertainty of 4.8 mBq, and an estimated bias of 3.7 mBq. The noted increase in percent measurement uncertainty with increasing sample activity is somewhat puzzling and unexpected. Two root causes have been discussed: a significant increase in isobaric interference with the synthetic urine; and the level of ^{242}Pu spike used in the analysis. The isobaric interferences have been linked to the possibility of undissolved salts being introduced into the mass spectrometer. The intercomparison results did indicate that the Laboratory's TIMS system is appropriate for application in the plutonium bioassay program.

Application of the same class-100 clean room radiochemistry and two-stage TIMS yielded an average measurement uncertainty of 0.4 mBq per 24 h \pm 100%. This measurement capability corresponds to a minimum detectable CEDE of 0.2 mSv. The results based on 12 urine samples indicate the two-stage TIMS has the capability of detecting lower amounts of plutonium than the single-stage TIMS. The chemical separation procedures have not been refined to the state that they can use the full detection capability of the two-stage TIMS. This is based on the fact that we cannot obtain the same dynamic range for chemically processed samples that we can for the direct loading of tracer. At the present time, two-stage TIMS is not suitable for routine bioassay analyses. However, we recommend that research and development continue to upgrade the two-stage TIMS capability.

Table 1. continued

ID	²³⁹ Pu urine concentration (μBq24h^{-1})	σ (μBq24h^{-1})	Analyzed volume	Collected volume	Plutonium work?
60	7.0	2.2	2330	2330	Y
61	19424.6	158.0	700	1675	Y
62	2563.0	41.1	700	1430	Y
63	41.8	35.2	700	2460	N
64	64.0	51.1	700	3270	N
65	300.8	13.7	700	775	Y
66	770.3	55.1	700	2715	Y
67	1.5	18.1	700	1340	N
68	-2.2	29.6	700	1950	N
69	20.4	22.9	500	1030	N
70	58.1	38.1	700	2235	N

Table 2. Summary of LANL bioassay techniques including the average measurement error, the associated classical MDA, and the minimum detectable dose at 180 days after an acute intake of inhalation class Y, μm AMAD ²³⁹Pu, for a single annual urine sample. The synthetic urine results are based on the NIST results. The theoretical limit for the two-stage TIMS is based on 12 human urine samples.

Method	σ_0 (μBq24h^{-1}) [atoms 24h ⁻¹]	MDA (μBq24h^{-1})	CEDE (mSv)
TIMS _{single stage} human urine	3.8 [4.2 x 10 ⁶]	17.5	1.9
TIMS _{single stage} synthetic urine	4.8 [5.3 x 10 ⁶]	22.1	2.5
TIMS _{two stage} human urine	0.4 [4.1 x 10 ⁵]	1.7	0.2
α -spec humane urine	150 [2 x 10 ⁸]	700	90

To test the influence of the lower measurement error, we compared the TIMS and alpha-spectroscopy techniques using distributions of random urine excretion patterns generated by a series of Monte Carlo experiments. In the first case, the assumed intake scenario was an acute inhalation of 12 Bq, $1\mu\text{m}$ activity-median aerodynamic diameter (AMAD), inhalation class Y, Pu²³⁹. It was further assumed in our analysis that urine samples were collected on days one, three, and five after the intake for the TIMS analysis. The samples from the TIMS protocol were generated with a measurement error of 3.81 mBq per 24-hour urine sample. Samples were collected on days one, two, four, eight, 16, 30, 60, 120, and 240 after the intake for the alpha-spectroscopy analysis. The samples from this second protocol were generated with a measurement uncertainty of 150 mBq per 24-hour urine sample. A biological variation of 10% was used for both data sets in conjunction with the ICRP Publication 30 lung model and the Jones plutonium excretion model in the Monte Carlo calculations to generate two sets of 100 random realizations of plutonium concentrations in urine due to the defined intake (Jones 1985, ICRP30 p1, Moss 1969). The two sets of random realizations were analyzed with a mathematical unfolding technique developed at Los Alamos by Miller et al. (Miller 1993, Miller 1995, Miller 1996a, Miller 1996b, Miller 1997).

The results of the two analyses are displayed in figure 4. The intake scenario would be expected to deliver a CEDE on the order of 1 mSv. Note the poor performance of the alpha-spectroscopy technique at this level. The results indicate a nearly 100% failure rate for detecting an acute intake resulting in 1-mSv CEDE, even if immediately followed up with an intensive nine sample urine assay program. The TIMS analysis reveals distribution of CEDE estimates with an expectation of 1 mSv and a population standard deviation of 0.1 mSv.

Monte Carlo calculations were also performed over a range of CEDEs (0.01 to 1000 mSv) for both types of urine analysis. In this case, results are based on

the collection of three 24-hour urine samples at days one, three, and five (for both measurement methods) after an inhalation intake of class Y, 1- μ m AMAD, ^{239}Pu . The biological variability used for both data sets was 10%.

Results presented in figure 5 clearly indicate the useful sensitivity of the TIMS measurement technique over alpha-spectroscopy. The TIMS results are accurate to committed effective doses down to the 0.2-mSv level, compared to 5 mSv for alpha-spectroscopy. In addition to the higher level of accuracy, the TIMS method also provides a remarkably low false negative fraction with respect to the US DOE's monitoring requirement of 1 mSv, as shown in figure 6. As seen in figure 7, the relative measurement errors provided by TIMS are also quite low when compared to alpha-spectroscopy. TIMS provides a coefficient of variation on the order of 10% at 1-mSv CEDE.

An interesting feature of the mass spectrometry technique is the capability to obtain information regarding the presence and concentration of both ^{239}Pu and ^{240}Pu (Perrin 1985, Efurd 1995b). The ratio of ^{240}Pu atom to ^{239}Pu atom content in a sample reveals some information about the source of the plutonium (Efurd 1995b). For example, material used in the fabrication of the World War II nuclear weapons had a ratio of 0.01, weapons material in current use has a ratio 0.06, and environmental material from open-air testing has a ratio of 0.18. This information is applicable in establishing the approximate era and source of intake. Preliminary results indicate that this technique will identify statistically significant ratios when ^{239}Pu urine concentrations exceed 400 mBq per 24-hour urine sample (approximately 5×10^8 atoms).

Application to monitoring workers.

The TIMS analysis was implemented as part of the LANL routine and special bioassay programs in January 1997. Individuals who handle plutonium are routinely monitored through the collection of an annual 24-hour urine sample. The sample is processed through the ultratrace chemistry procedure in class-

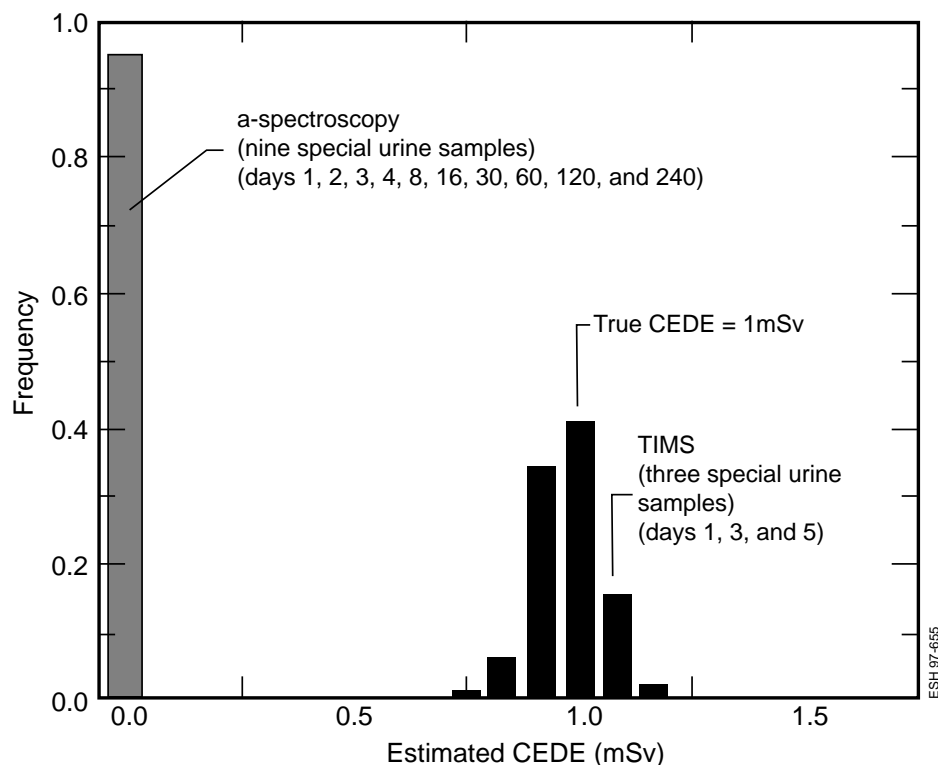


Figure 4. Distributions from two 100-trial Monte Carlo experiment. Both distributions are based on an inhalation of 12Bq, inhalation class Y, 1 μ m AMD, ^{239}Pu . This intake corresponds to CEDE of 1 mSv. Results are based on an inhalation intake of class Y, 1 μ m AMD, ^{239}Pu . The biological variability used for both data sets was 10%.

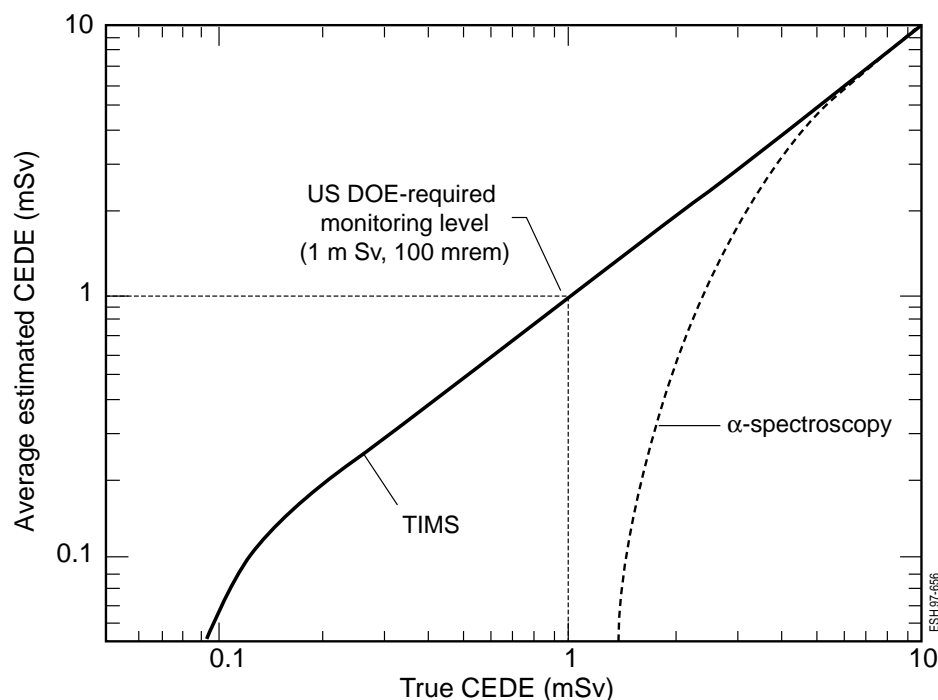


Figure 5. Monte Carlo average estimated CECE for the indicated true CEDE for α -spectroscopy and TIMS. Results are based on the collection of three 24-hour urine samples at days 1, 3, and 5 after an inhalation intake of class Y, 1 μ m AMAD, ^{239}Pu . The biological variability used for both data sets was 10%.

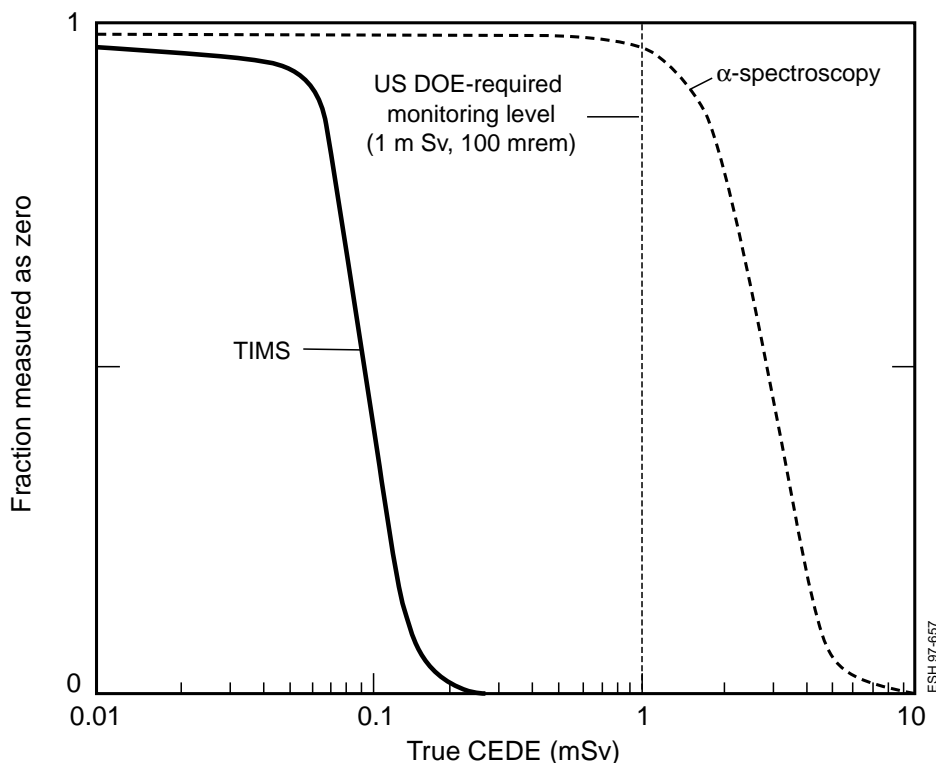


Figure 6. Monte Carlo estimated fraction of intakes determined as zero intake for the indicated true CEDE for α -spectroscopy and TIMS. Results are based on the collection of three 24-hour urine samples at days 1, 3, and 5 after an inhalation intake of class Y, $1\ \mu\text{m}$ AMAD, ^{239}Pu . The biological variability used for both data sets 10%.

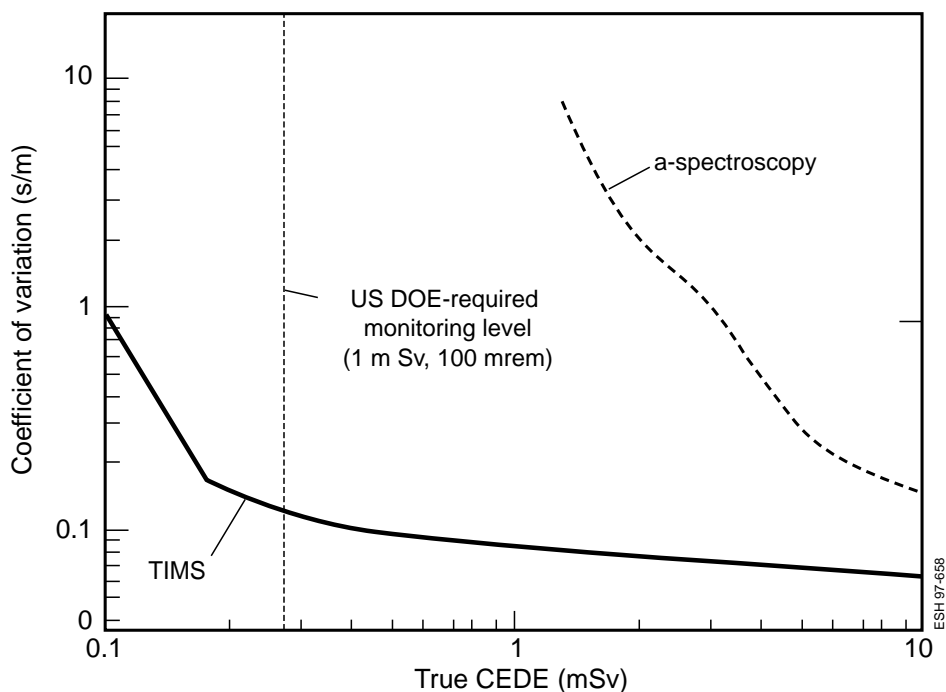


Figure 7. Monte Carlo estimated relative error for average estimated intakes for the indicated true CEDE for α -spectroscopy and TIMS. Results are based on the collection of three 24-hour urine samples at days 1, 3, and 5 after an inhalation intake of class Y, $1\ \mu\text{m}$ AMAD, ^{239}Pu . The biological variability used for both data sets 10%.

100 clean rooms and then is analyzed by both alpha-spectroscopy and TIMS. Use of the alpha-spectroscopy allows for a direct measure of chemical efficiency and the detection of ^{238}Pu in the urine sample. The ability to detect committed effective doses in the 1- to 3-mSv range from routine samples in the year of intake will be in place in 1998 when the population and individual baselines are established based on 1997 routine results. Detection of intakes in the 10- to 20-Bq range is currently possible under the special sampling program for known exposure situations. For situations in which the committed effective dose is expected to exceed 10 mSv, three 24-hour urine samples are collected over the five days following the exposure situation. Additional samples may be collected at later times if necessary. For cases where the committed effective dose is expected to exceed 1 mSv, a single 24-hour urine sample is collected following the exposure. Additional samples may be collected at later times if necessary. The current turn around time to assess intakes from such exposure situations is three to six weeks. In emergency cases where the CEDE is expected to exceed 100 to 250 mSv, analysis may be performed in three to seven days.

The following summarizes study results:

- The significance of lowering measurement errors relative to routine detection of intakes results in a detectable CEDE on the order of 3 mSv.
- The combination of ultratrace chemistry performed in class-100 clean rooms and detection by TIMS produces a superior bioassay result.
- Distributional analysis shows a relatively small bias and a low false negative rate when TIMS is applied to special bioassay.
- The availability of a high-quality estimate of the CEDE in combination with fewer samples should provide the framework for better work place decision making, optimized monitoring of cost benefit, and improvements in the ability to demonstrate strict compliance with DOE regulations.

Deliverables

The following lists project deliverables and their status:

- A method for detecting low concentrations of ^{239}Pu in urine from occupational sources.

Status: Complete. The project was awarded a 1996 LANL Distinguished Performance Award.

- A method for detecting low concentrations of ^{239}Pu in urine from environmental sources.

Status: Synthetic urine studies complete. Two-stage measurements complete. Electronics upgrade complete. Intercomparison study complete.

- Statistical distribution information for application of Bayesian techniques.

Status: Complete

- Participation in DOE's Laboratory Accreditation Program (DOELAP) certification process.

Status: Currently participating in the DOELAP process. Discussions are underway to prepare appropriate DOELAP. We are in the process of preparing DOELAP applications and have begun a project to complete program documentation. The initial internal audit is underway.

- Contacts with DOE, Defense Nuclear Agency (DNA), and other potential funding agencies.

Status: Participating in the DOE OIHS intercomparison. Chemistry and measurements are complete. The final report has been submitted to DOE. DOE is currently publishing results.x

Status: Results were presented at 1996 Annual Meeting of Health Physics Society and at 1996 the Annual Meeting on Bioassay, and Environmental Radiochemistry. Manuscripts are in preparation for publication in the open literature.

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High-Energy Neutron Dosimetry and Spectroscopy

Principal Investigators: Robert T. Devine, Health Physics Measurements Group (ESH-4); L. Scott Walker, Health Physics Operations (ESH-1), and Hsiao-Hua-Hsu (ESH-4)

Funding: FY95, \$14K; FY96, \$14K; FY97, \$23K. The Neutron Science and Technology Group (P-23) has donated six weeks of beam time for each year of this project. The ordinary charge rate is about \$300 to \$500 per hour.

Introduction

Traditionally, personnel protection at high-intensity accelerators has been based on shielding. Upgrades in beam power—and the resulting potential for configurations through which certain equipment or procedural failures can send a high-intensity beam to an area that normally receives a low-intensity beam—have led to the use of beam-limiting and radiation-limiting interlocks. The reason for this is that shielding under these conditions would become prohibitively expensive. To complement a system of interlocks, a high-dose measurement capability is needed. The proposed use of Los Alamos Neutron Science Center (LANSCE) as a spallation source for accelerator production of isotopes and tritium production makes this capability particularly relevant to our study.

Physicists need the application of current technologies to monitor fields so that a high-dose measurement response can be obtained for personnel who are involved in survey work and in operations that have a higher risk of delivering a dose. Such devices have not been calibrated in the high-energy fields at Los Alamos National Laboratory (LANL, Laboratory), but some studies of the current technologies indicate that they will respond significantly in high-energy fields. The preliminary studies of the use of track-etch dosimeters in lead spheres can also allow either a form of area monitor or some increase in the area of applicability of a type-Bonner sphere spectrometry.

With the information arising from this study, LANL could make a proposal for the development of a calibration facility that could obtain \$250K in support the

Department of Energy (DOE) every three years. Attracting these funds would be the benefit derived from doing a pilot intercomparison program using the current white neutron fields at (WNR). We need to do some preliminary work in increasing the field size and possibly improving the purity of the spectrum, if the service is to be offered on an extended basis outside the DOE complex.

The ultimate objective is to provide neutron fields for calibration that could be integrated into the fields currently available from *Physikalisch-Technische Bundesanstalt*, which currently go from thermal to 20 MeV and may be extended using the Louvain accelerator at powers up to 100 MeV. There are fields which are well determined at the European Center for Nuclear Research but are of energy greater than 1 GeV. A facility, such as LANSCE, which covers the range from 100 MeV to 1 GeV, covers this needed energy interval.

Method

We used fields produced by a spallation target struck by a beam of 800-MeV protons at the 15 left port of LANSCE. The fields were filtered by 0-, 4-, and 16-inches of polyethylene placed between the target and the experiment. We calculated the corresponding spectra using Monte Carlo methods. The track-etch dosimeters were exposed on a polyethylene phantom. The dosimeters were exposed bare and behind filters of 0.1-, 0.2-, 0.3-, 0.4-, 0.5-, and 0.6-inches of lead with the face of the dosimeter perpendicular to the beam. To study the angular dependence of the response, we exposed the dosimeters bare and behind filters of 0.2-, 0.4-, and 0.6-inches of lead with the face of the dosimeter at 45° and 75° with respect to the incoming beam.

We performed Monte Carlo calculations of these experimental configurations using MCNP and LAHET with input from the calculated spectra. These results are compared with the experimental results to understand the basic processes involved in the production of tracks with high-energy neutrons.

Progress and results

The experimental results are shown in the following three figures.

Our Monte Carlo calculations are preliminary, but the results when normalized to 0° for each spectrum appear to follow the same trend as the experimental results. The final results will be completed soon, but the time required for each calculation is significant at higher energies.

A proposal for a high-energy neutron dosimetry intercomparison was submitted to DOE EH 41, May 14, 1997. Uncertainties in the DOE budget have not allowed any final decision to be made on the funding.

Conclusion and deliverables

The 0-inch polyethylene filter results have a considerable angular dependence while the 4- and 16-inch filters show almost no angular dependence. The method used for fission neutrons to mitigate this effect is to place the dosimeters on the face of a triangular pyramid with the base facing the body and the faces at a 35° to the base. This does not flatten the response but brings it into line with the expected change in dose as the body is rotated with respect to a parallel beam directed from anterior to posterior. A flat response would overestimate the true response. The response of the dosimeter decreases with increasing

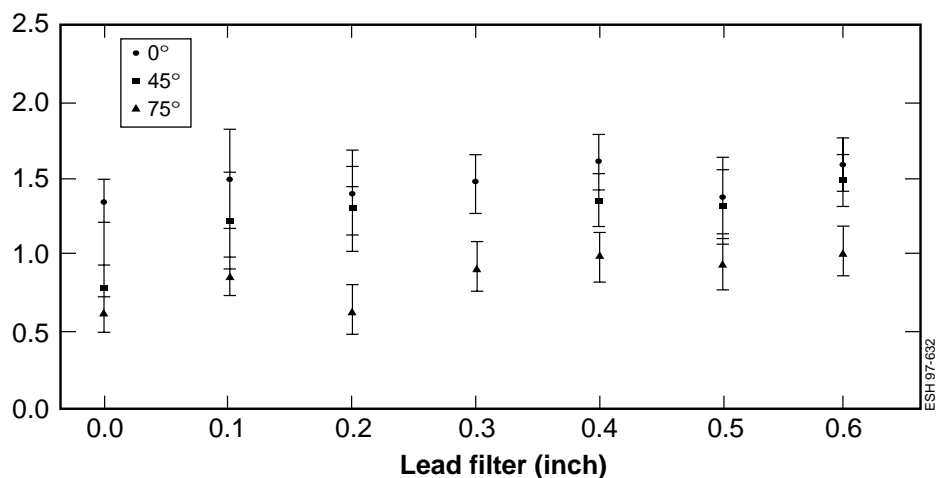


Figure 1. 0-inches polyethylene in beam

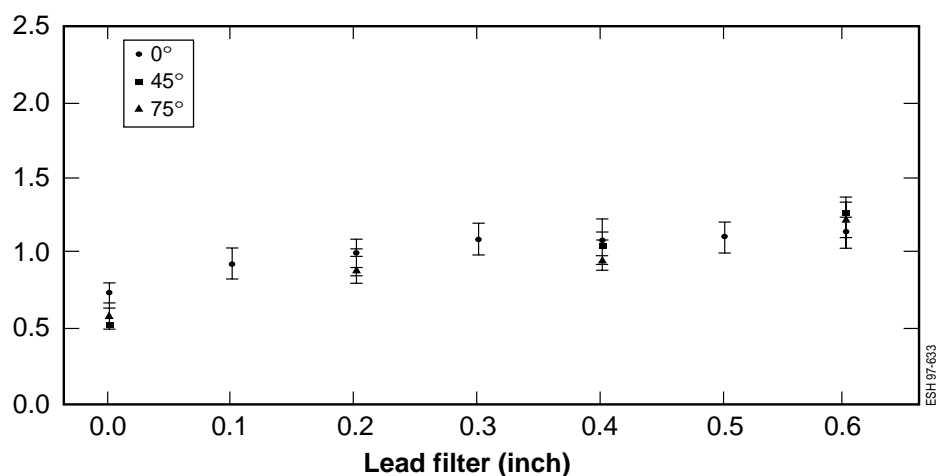


Figure 2. 4-inches polyethylene in beam

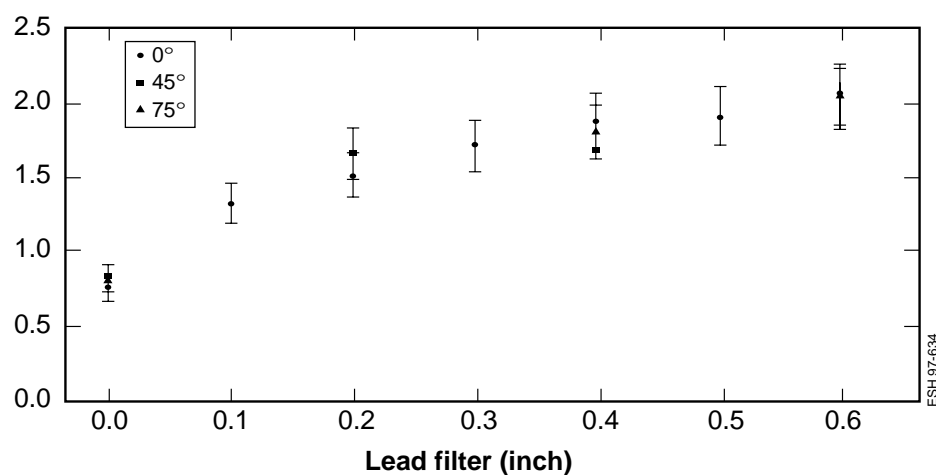


Figure 3. 16-inches polyethylene in beam

energy, but the onset of the $Pb(n,xn)$ reaction increases the number of neutrons the dosimeter is exposed to and therefore the number of tracks. At this writing, a thickness of 0.2-inches lead would seem to give a reasonably flat response over the energy range.

A cube with six track-etch dosimeters, one on each face with 0.2-inches lead sheet in front and in back of each face will be constructed and exposed in the next cycle at LANSCE.

Development and Implementation of the Los Alamos National Laboratory (LANL) Neutron Extremity Dosimeter

Principal Investigators: M. W. Mallett, J. M. Hoffman, Health Physics Measurements (ESH-4); A. R. Wood-Zika (Col), Department of Nuclear Engineering, Texas A&M University

Funding Level: FY97, \$13K

Introduction

Radiation-induced damage to the human body was first observed as skin burns on hands that had been repeatedly exposed to x-rays. In the workplace, preventing related health injuries, such as those shown in figure 1, is the objective of personnel extremity dosimetry. Current regulations limit the total equivalent dose to the extremities to 0.5 Sv annually. At LANL, neutron exposure, in particular, represents one component of the extremity dose for glove-box workers. Prior to the effort represented in this study, no effective technique has been developed for measuring the neutron extremity dose. This is because the relatively small amount of tissue mass present in the hands represents an obstacle to using traditional neutron sensitive



Figure 1. Example extremity injuries following acute radiation exposure.

thermosluminescent dosimetry (TLD) materials for measuring neutron extremity dose.

At LANL, the neutron extremity dose has historically been reported as equal to the measured gamma extremity dose. Measurements of glove-box operations at LANL contradict this 1:1 neutron-to-gamma dose ratio. However, individual-specific neutron-to-gamma extremity dose ratios are impractical owing to the large number of field measurements such a solution would imply. Recent developments in high-energy neutron dosimetry made by the Measurements Technology Support and Personnel Dosimetry Operations teams of ESH-4 have been used to develop a new extremity dosimeter that accurately measures the neutron extremity dose without the need for extensive field measurements.

Progress and results

Dosimeter. A neutron extremity dosimeter was designed to be worn on the wrist of glove-box workers, as shown in figure 2. The dosimeter holder was manufactured locally¹ and is constructed of a canvas gortex material.² Three pilot issues of the new dosimeter were conducted at technical area 55. Suggestions from workers resulted in design modifications to facilitate donning and doffing ease, including the incorporation of a metal snap-on wrist band and a velcro fastener.

The dosimeter contains a PN3 detector³, a CR-39 foil that exhibits relatively flat energy response over the energy range 0.2–20 MeV. The dosimeter also includes two ⁶LiF and two ⁷LiF TLD phosphors, one of each type shielded by a 0.16-mm-thick cadmium



Figure 2. A properly donned LANL neutron extremity dosimeter.

filter. The ⁷LiF phosphor pair measure the gamma extremity dose received by the worker. This quantity is subtracted from the ⁶LiF phosphor response to determine the net neutron signal. The ⁶LiF phosphor pair are particularly sensitive to thermal energy neutrons and because of the cadmium filter, provide a relative measure of the neutron energy spectrum.

Laboratory calibration of the dosimeter was performed using a National Institutes of Standards and Technology (NIST) NIST-traceable ²⁵²Cf source. Polyethylene spheres ranging in diameter from 0–20 cm were used to moderate the source, thereby increasing the thermal energy component of the neutron spectra by as much as a factor of 10. Also, the moderation effectively reduced the average neutron energy from 2.54 MeV to 1.15 MeV.

Figure 3 displays the dosimeter characterization results. These data show that the PN3 detector measures very near the true neutron dose for neutron spectra with a minimal thermal energy component. However, as the thermal energy component increases, the PN3 detector underresponds the true neutron dose.

As expected, the cadmium-covered TLD-600 response increased with decreasing average neutron energy (i.e., an increased thermal energy component). In addition, the ratio of the bare TLD-600 response to the cadmium-covered TLD-600 response increased as the thermal energy component of the neutron spectra grew. Hence, the ratio of the TLD-600 responses can be used as a neutron energy spectrometer. As a result, the reported neutron extremity dose can be calculated as the sum of the PN3 measured neutron dose plus the cadmium covered TLD-600 measured neutron dose adjusted by an appropriate weighting factor based on the bare-to-cadmium-covered TLD-600 ratio.

Dose analysis. Accurate reporting of the neutron extremity dose requires the application of a suitable dose equivalent conversion factor. Current neutron fluence-to-dose equivalent conversion factors reported in National Council on Radiation Protection's (NCRP's) Report No. 38 were derived using a 30-cm diameter cylindrical torso phantom. Hence, these values are not directly applicable to extremity dose calculation.

As a part of this project, extremity-specific neutron fluence-to-dose equivalent conversion factors were computed via Monte Carlo techniques. Several forms of extremity phantoms were modeled for these calculations including finger and arm phantoms previously reported by Department of Energy Laboratory Accreditation Program (DOELAP) and PNNL, as well as the arm/shoulder phantom⁴ used for calibration exposures at LANL. The results indicated less than a 10% difference between extremity-specific and whole-body neutron fluence-to-dose equivalent conversion factors as reported in NCRP Report No. 38. However, the NCRP values were established using quality factors based upon the risk of exposing the blood forming and other major organs of the torso. The development of extremity-specific neutron quality factors would necessitate extensive biological research beyond the scope of this project.

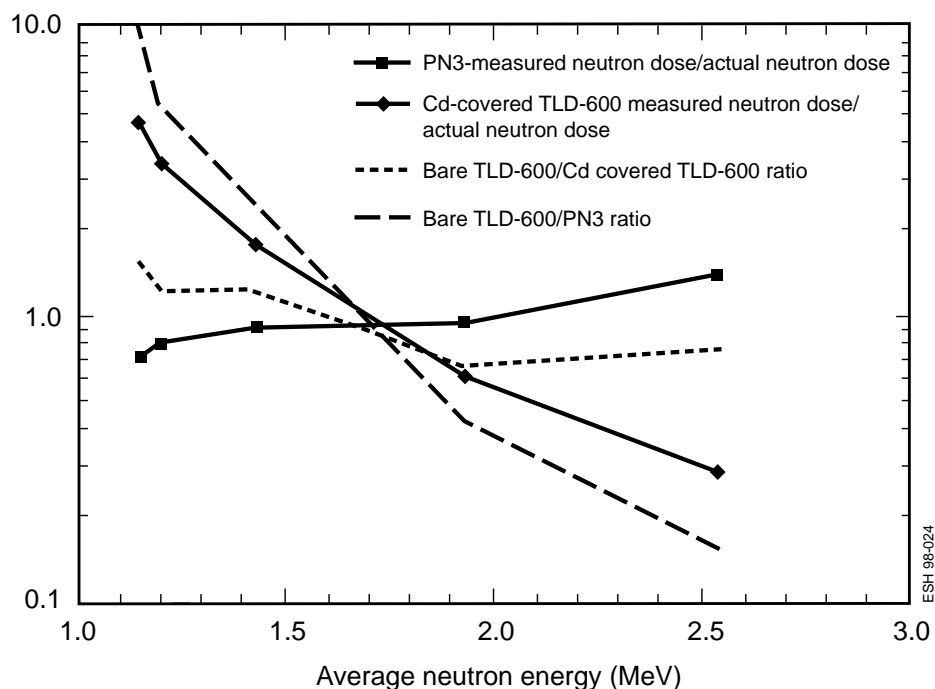


Figure 3. Calibration results for the LANL neutron extremity dosimeter using polyethylene-moderated ²⁵²Cf.

Deliverables

The deliverable for this project is the accredited use of the Laboratory's neutron extremity dosimeter as an integral part of the LANL external dosimetry program administered by ESH-4. The new dosimeter is currently being submitted for accreditation by DOELAP. Blind testing of the dosimeter will commence during July 1998. Additional effort will also be made to determine appropriate neutron fluence-to-dose equivalent conversion factors applicable to the extremities. This will include extensive peer review of the calculations reported here coupled with the pursuit of initiating an American National Standards Institute working group for standard development.

¹ Currently: Arizona Sewing Contractors, Glendale, AZ.

² FRHAM Safety Products, Inc., 318 Hill Avenue, Nashville, TN 37210.

³ Bicon-NE, 6801 Cochran Road, Solon, OH 44139.

⁴ Radiology Support Devices, 1904 E. Dominguez Street, Long Beach, CA 90810.

Optimization of Continuous Air-Monitoring (CAM) Instrument Placement

Principal Investigators: J.C. Rodgers, J.J. Whicker, Y. Yang, M.E. Moore, H. Gong, Health Physics Measurements (ESH-4); J. Spore, Nuclear Systems Design and Analysis (TSA-10); R. Sripsick, Industrial Hygiene (ESH-5)

Funding: FY95, \$100K; FY96, \$98K; FY 97 \$77K. Additional support for the project was obtained from Nuclear Materials Technology Division during FY94 (\$50K) and \$110K was obtained from the Capabilities, Maintenance, and Improvement Project during FY96.

Introduction

Continuous Air Monitors (CAMs) provide real-time measurements of radioactive aerosol concentrations and exposures and are designed to alarm when the exposure (DAC-hr) exceeds an alarm threshold. Within a plutonium nuclear facility, alpha CAMs are among the engineered safety features that limit and reduce worker exposure (and dose) should loss of containment occur. The effectiveness of CAMs is generally limited by sensitivity and timeliness of response. Effectiveness can be degraded by poor instrument design, improper maintenance, and inadequate planning in their utilization.

Dramatic improvements in alpha CAM technology have led to the instrument's greater sensitivity over the past decade (McFarland et al. 1992); however, somewhat less attention has been given to the issue of their optimized utilization. A number of studies designed to evaluate the impact of CAM placement on the timely detection of airborne releases in plutonium facility laboratories have been conducted (Fairchild et al. 1991, Sripsick et al. 1979, and Whicker et al. 1997). These studies found that the placement of CAMs can have a significant influence on how effectively and quickly CAM alarms are generated following a release. They support the findings of the multifacility study by Crites (1994) that shows inadequate alpha-CAM utilization in DOE plutonium facilities can often lead to exposures greater than 500 DAC-hrs without detection. The implication of these studies is clearly that a process of well-considered placement and utilization of alpha CAMs is an essential component of planning for optimized worker protection

in a plutonium facility (International Commission on Radiation Protection [ICRP] 1991).

The facility installation and maintenance costs for a CAM system can be significant. The largest of these costs are fixed: base facility costs associated with design and installation of utilities needed for CAMs (power, communication, vacuum) and costs associated with routine maintenance and calibration of radiation protection instruments. Both do not change appreciably by small incremental changes in the number of CAMs installed. Other costs are incremental and associated with adding (or deleting) the number of utilized CAMs. Of course, there can be very large incremental costs, amortized over the life of the instruments, which are associated with a facilitywide upgrade of instrumentation (and perhaps associated utility requirements). When one considers costs in optimization, however, the overall cost increments or decrements associated with increased worker protection (or intervention, in ICRP terms) must take into account the incremental monetary cost of installation and maintenance and also the potential cost savings associated with decreased worker dose (i.e., \$10k per person-rem). Similarly our consideration must include institutional costs (e.g., facility down time and investigations) associated with accidental releases that result in high worker exposures that could be in the hundreds of thousands of dollars per week.

Sripsick et al. (1979) studied optimized CAM placement for protracted releases (i.e., > 900 seconds) and made concentration measurements under steady-state conditions and integrated over periods longer than the releases.

They found that the level of worker protection increased as the number of CAMs increased, but that the rate of the increase in protection asymptotically reached zero. After the inclusion of a certain number of CAMs no further benefit in worker protection was achieved. Merwin et al. (1989) used the data from Sripsick et al. (1979) and considered the cost of CAMs and dose savings for CAM use in an example of optimizing the number of CAMs. The conclusions were again constrained to the aforementioned experimental conditions; yet, there is evidence that short-duration (puff) releases are much more common than continuous releases. For example, McAtee (1990) provided historical evidence that showed that most releases in the Plutonium Facility at Los Alamos National Laboratory (PF-4) were puff releases. The dispersion and worker exposure dynamics for puff releases can be quite different from protracted releases. For example, puff releases create a highly concentrated cloud around the release point; the cloud then dissipates into the rest of the room. This situation creates a situation in which a worker collocated with the release can be initially exposed to a highly concentrated aerosol that cannot be detected by CAMs located elsewhere in the room and by sampling outside the cloud. Mixing of the aerosol in the room then disperses the aerosol until it is detected by a nearby CAM.

Puff releases present a difficult but common challenge for CAM detection and optimized placement. To determine optimized placement under the condition of a puff release, it is important to collect aerosol concentration data more finely resolved in time and space than in many

past studies (Scripsick et al. 1977; Brunskill and Holt 1967; Brunskill and Hermiston 1966, Vavasseur et al. 1986). The resolved data would enable investigations of the dynamics between potential CAM response and worker dose under conditions of transient puff releases.

To develop strategies for optimal CAM placement, we undertook tracer particle studies in several plutonium laboratories in PF-4 and in an experimental test room. We also performed computational fluid dynamics (CFD) modeling of these spaces to determine if CFD modeling could be used as a reliable alternative to field investigations for CAM optimization. The purpose of these studies was to determine a method to optimize placement of CAMs and to explore the uncertainties of placement decisions.

Method

Field studies. A series of test aerosol releases were conducted in PF-4, rooms 209 and 420. To simulate a plutonium release, we used an orifice nozzle generator to inject polydisperse oil (dioctyl sebacate) test aerosols into the rooms from various locations. A network of twelve or more laser particle counters (LPCs) recorded concentrations at a variety of sampling locations every 10 seconds. Therefore, the dispersion of the aerosol could be measured resolved in time and space. Each release simulated a glove-box glove release and so the release locations were about 15 cm from the glove-box face. One LPC recorded the breathing zone concentrations at the release location and was used to estimate worker exposure. The use of this LPC allowed us to compare simulated worker exposure and CAM response. Figure 1 illustrates the release and sampling locations for each of the rooms.

We used several techniques to analyze for optimized placement of CAMs based on the data collected with LPCs during test aerosol releases in rooms 209 and 420 in PF-4. The first analyzed the relationships between possible dose savings (DS) and the number and

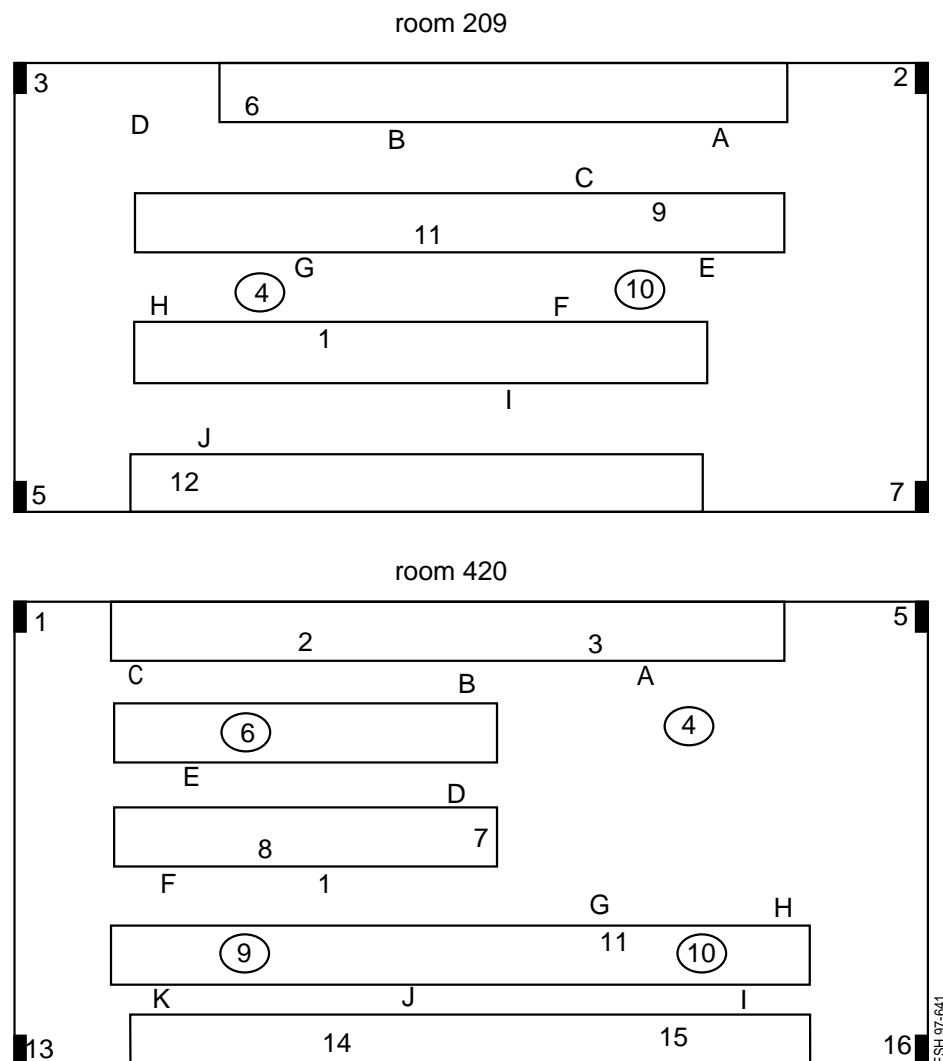


Figure 1. Approximate position of sample, release, and ventilation supply and exhaust locations in test rooms. Numbers represent sampling positions and letters represent release positions. Open rectangles are glove-box lines and ovals indicate position of supply diffusers on the ceiling. Wall-mounted, floor-level exhaust registers are shown by the shaded rectangles in the corners.

placement of CAMs by studying the interplay between simulated worker exposure and CAM response. Using data from PF-4, we developed a DS metric that simply defined was the amount of reduced dose given an arrangement of CAMs in the room. Mathematically, the DS was one minus the ratio of the exposure (time-integrated concentration in the breathing zone of a simulated worker) under the condition where they were protected by alarming CAMs, divided by the exposure if there were no CAMs in the room. The effect of CAM instrument response time on the DS was also explored.

Optimized CAM placement and the associated uncertainty was determined by analyzing the DS at the discrete sampling locations for all releases. We determined the best combination of sampling locations for a given number of CAMs by calculating the highest mean DS as averaged over all release points. The uncertainty in DS was calculated for each sampler and for all release locations. Multiple measurements determined this uncertainty which is expressed as the standard error of the mean DS.

Then, we performed a spatial analysis using the geographical informa-

tion and decision-making software IDRISI to use unsampled locations in the optimization decisions. Specifically, the spatial analysis was used to provide probability boundaries for any established DS goal or threshold for worker protection in the room.

Another method that possibly extends the results of DS analysis is that we consider that both rapid response (short lag time) and interception of high concentrations of aerosol before dispersal are desirable attributes of a monitoring location. Then a figure of merit (FOM) that includes lag time, T_L , and the ratio of peak to average concentration, C_M / C_A , (a measure of the peak shape) would provide a ranking of locations. The ratio $FOM = T_L / (C_M / C_A)$ provides a ranking of desirable CAM locations that might help extend the DS assessment. A great advantage of the FOM approach to optimized CAM placement is in its ease of calculation and use. It has been found that the DS method and FOM ranking track very closely except in those cases where the monitored location is very close to a release. In this case, the peak to average ratio is large, which creates a very low FOM. Other methods, in addition to these, for incorporating peak shape in the optimization process are under investigation.

Experimental test room. Several experimental studies were conducted in the experimental room at the UHTREX location. We undertook one of the studies to attain additional validation of computational fluid dynamics (CFD) code predictions. For this, we used a 3-D sonic anemometer to make both aerosol dispersion and air flow measurements. The results of this validation work are still being analyzed.

We also undertook other studies to explore differences, as a function of particle size and electrical charge, in aerosol dispersion and removal rates after a release. Optical particle counters such as the LPCs and the MET ONE particle sizer were used to measure particle concentrations categorized by particle size. The results of these studies were important to our achieving a fundamental

understanding of the dispersion and fate of particles once they were injected into room air streams. We found this information useful to achieve a better understanding of CAM response to releases, to understand surface contamination distributions in rooms after releases, and to determine important aerosol characteristics for use in quantitative air flow studies for CAM placement.

Computational modeling. Beyond the already mentioned validation investigations, during the past year we undertook an extensive analysis of the CFD code CFX 4.1 and used the code to model room air flows and particle trajectories for a actual plutonium aerosol release occurring in room 3111 in the CMR building (Rodgers et al. 1997). For this model, particle motion was assumed to be affected by Stokes drag, turbulent diffusion, and gravitational settling. Two different particle sizes, 1 mm and 10 mm, were used in the calculations.

Progress and results

Our data show that optimized placement of CAMs, when compared to the exhaust register placement strategy currently used, could significantly improve worker protection and at less cost. We recognized that in addition to optimizing CAM location and utilization, enhanced ventilation air flow patterns may also contribute to dose savings. Therefore, we explored alternative ventilation designs using CFD modeling. Model predictions suggested that improvements in worker protection could be achieved by directing air flow downward at work stations and toward optimally located CAM instrumentation, but much work remains to be done in this area.

Using concentration measurements collected from PF-4, rooms 209 and 420, we found that placement of CAMs at interior locations and away from the corner registers would probably result in more sensitive and faster detection of Pu aerosol releases (Whicker et al. 1997). Specifically, we found that the average dilution of aerosol among all release locations and individual sampling

locations was generally less when the sampling locations were toward the interior spaces of the rooms. Also, we found placement of CAMs at interior room locations would also result in faster alarms because lag times (time for the aerosol to travel from the release location to the sampling location) were significantly less at interior room locations than those at the exhaust registers. These results would likely be true whether the release was a large puff release (type assumed for our experiments) or a low-level chronic release. In combination, the lag time and time profiles of the concentration for each release and sampling combination contributed to the evaluation of optimized placement because each was used to calculate the DS metric.

Figure 2 shows the relationship between DS and the number of optimized samplers (combination of samplers with highest DS for a given number of samplers) under differing instrument response times, based on data from room 209. This figure shows a plateau in computed DS at about three or four LPCs (surrogate CAMs). That is, there is an optimal number of CAMs beyond which the potential dose savings do not justify the cost of purchasing additional CAMs and their associated maintenance. It also shows the decrease in DS as the CAM instrument response time increases. These results were also found for room 420. Although the experimental designs were different, these findings are similar to prior studies by Scripsick et al. (1979).

Figure 3 shows an optimized placement strategy for CAMs room 209 after geographic information system (GIS) analysis. The conclusions of this analysis are limited to some degree by the experimental constraints of having a finite number of samplers and release locations when an infinite number of possibilities for these exist. It would be useful to investigate the effects of the number and location of test releases and sampling locations on optimized CAM placement. To extrapolate the conclusions of the study beyond the experimental assumptions and limitations, we recommend that flexibility in CAM placement be maintained to achieve even

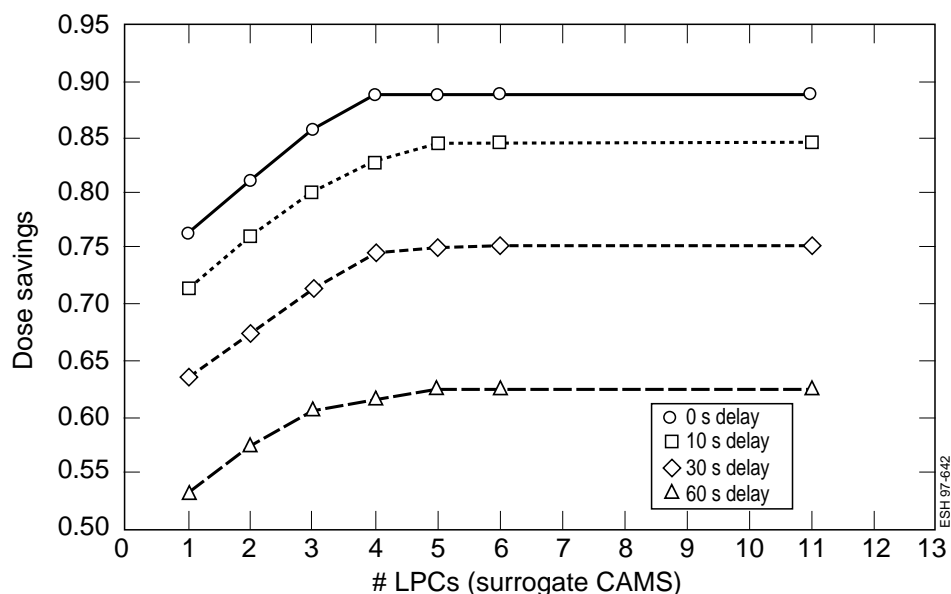


Figure 2. Fractional dose savings as a function of the number of samplers for the expected range of instrument response times. (Data from room 209)

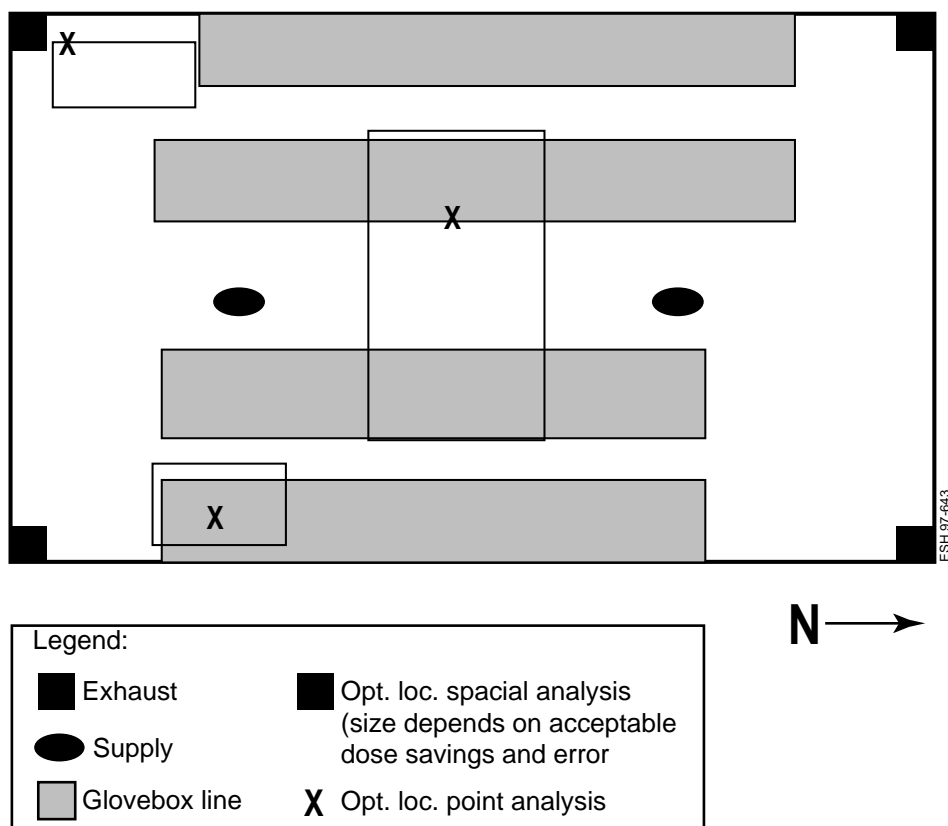


Figure 3. Optimal CAM placement for room 209

greater DS. For example, in the experiment we largely assumed random release locations which may not always be the case based on dynamic work patterns. Therefore, the optimal placement strategy is likely to have fixed CAMs, to cover work under the general condition where work is conducted throughout the room, combined with portable CAMs that can be moved to locations where work, especially more hazardous work, is conducted over longer periods.

We also explored the time dependency of the dilution factor (DF), which has historically been defined as the ratio of the breathing zone concentration to the concentration at some sampling location elsewhere in the room. Past studies showed that the DF was typically less than 10 and only rarely greater than 100. However, these measurements were based on protracted (over an entire sampling day) concentration measurements, which artificially lowers the DF for puff releases. Contrary to these results, we found that when puff releases were viewed as a time-varying factor, very large DFs existed and that they were especially large (i.e., >1000) during the first stages of the release (figure 4).

Finally, several personnel were trained in the CFD code CFX 4.1c. The ESH-4 SUN workstation was upgraded to Solaris 2.5 to run this CFD software. Simulation of air flows in the CMR building with CFX4.1c also suggested that problems regarding CAM placement exist in these laboratories. Modeling airflow patterns with CFX 4.1c suggested that there was a general movement of air from the ceiling to the floor, implying that there might be a benefit to move CAMs from the top of the slot-box to positions below the slot. However, confirmatory measurements of model input parameters are needed before any definite conclusions can be made. Also, as we expected, this simulation found that larger particles (10 μm aerodynamic diameter) deposited on the floor more rapidly than 1- μm -aerodynamic-diameter particles.

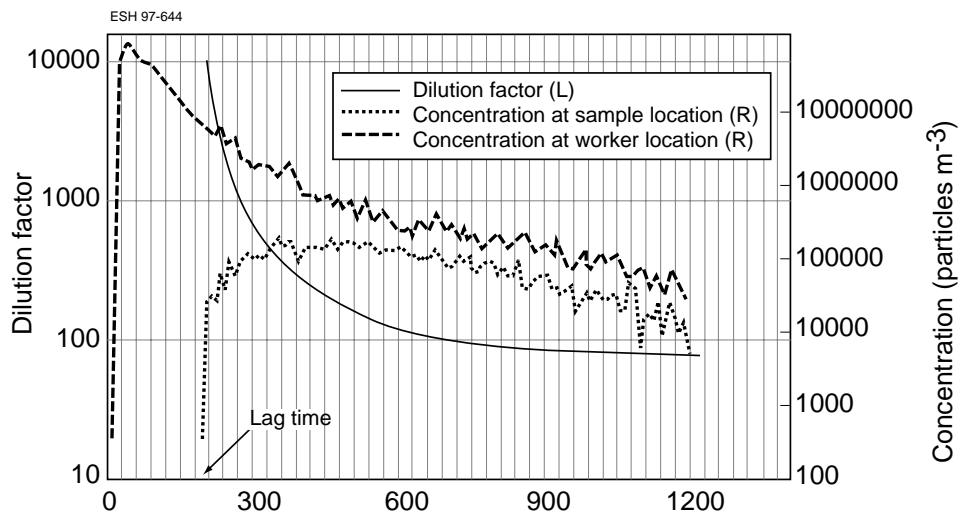


Figure 4. Concentrations for worker and sample locations and the corresponding dilution factor over time for a release in room 420.

Conclusions and deliverables

We find that significant reductions in worker risk from large inhalation exposures can be achieved through optimized placement of CAMs and that the cost of upgrading to the new generation of CAMs could be significantly reduced. One of the objectives of the project is that it provide a training document that describes the method for optimized placement of CAMs. This training document for sampling of transuranic aerosols has been written and published (McFarland et al. 1997). This document covers CAM use in a broad sense and includes a large section describing the CAM placement methodologies we have developed through this study.

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Resuspension of ^{238}Pu from Surfaces

Principal Investigator: Brian Rees, Health Physics Operations Group (ESH-1); Mary Ann Reimus (Col) Actinide Ceramics and Fabrication Group (NMT-9)

Funding: FY97, \$21K

Introduction

The high specific activity and specific energy emission of ^{238}Pu result in an apparent "resuspension" of its particulate contamination. The resuspension of ^{238}Pu results in the loss of control of highly respirable (less than $10\ \mu\text{m}$) particles. The resuspended particles cause unexpected contamination of small areas and personnel have the potential for inhalation, resulting in significant radiation dose.

The goal of this proposed study is to better understand the mechanisms of ^{238}Pu resuspension and to establish factors and principles to be used for more effective control of ^{238}Pu contamination. The resuspension of particles not only complicates contamination control, but may complicate ventilation clearance of rooms following a release. The elimination of airborne ^{238}Pu contamination may not be accurately described by standard ventilation exchange and clearance equations due to resuspension and kinetics from the high specific energy of ^{238}Pu . Excessive delay time before room reentry has an adverse impact on program deadlines and programmatic activities; premature entry may result in internal intake and unwarranted radiation exposures.

The resuspension of plutonium in transuranic wastes may also affect the rate of hydrogen production from hydrolysis of polymer (plastic) wastes by migrating to new, less hydrogen-depleted regions. Hydrogen production is an issue with about 20% of the wastes awaiting shipment to the Waste Isolation Pilot Plant.

An understanding of the mechanism of ^{238}Pu resuspension will improve the measures taken to protect workers and the environment from unnecessary

exposure to ^{238}Pu . With the small margin that exists between exposure and overexposure, improved control will have a significant impact on safety. Improved control will reduce the quantity of radioactive waste generated during work with ^{238}Pu . The elimination of $3\ \text{ft}^3$ of transuranic waste, one prevented intake, or two avoided skin contaminations is enough to offset the cost of this project.

Method

Planned. The basic experimental plan was to expose a variety of 1cm^2 test samples to a ^{238}Pu aerosol environment (glove-box interior), remove the samples from the exposing glove-box, and examine the loss of ^{238}Pu from these test samples. The activity of each sample was to be initially and then periodically recorded to track material loss. The test samples were to be separated to minimize deposition from adjacent samples and placed in a tray of ethylene glycol to minimize the potential for resuspension. The ethylene glycol was to be examined to evaluate the resuspended particles. Particles lost from the samples were to be assumed to be resuspended. The loss rate from different materials would help us to understand the mechanism(s) involved.

Materials were to be selected that have two characteristics:

- they are commonly used, and
- they represent a variety that separates thermal, physical, and electrostatic effects.

There was wide variety in the thermal and electrical conductivity of the test samples, as well as surface roughness. This experiment was designed to rapidly quantify ^{238}Pu resuspension rates from various materials. From these data, we expected that the mechanisms of resuspension would become clear.

The first step was to determine deposition rate to establish exposure time by exposing open top vials to a glove-box atmosphere and by examining the amount of material deposited. Then, shop-fabricated jigs were to be used to hold 25 test samples for exposure to the glove box environment. Twelve holding jigs were anticipated for a variety of experiments. The holding jigs allow consistent geometry, ease of handling, and separation of the samples from each other, while keeping samples above the ethylene-glycol level. We envisioned that the holding jigs and samples would be placed in petri dishes as shown in figure 1.

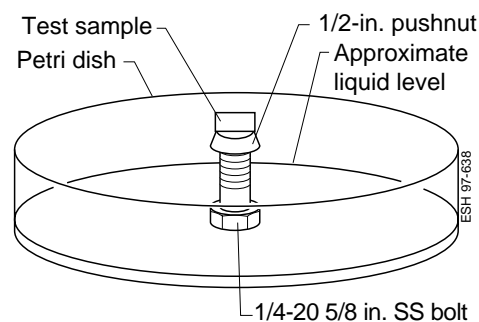


Figure 1. Schematic drawing of petri dish apparatus.

The loaded jigs were to be placed horizontally, vertically, and horizontally upside down. Various airflows to be directed across samples, both when the samples are stationary and when they are rotating on a turntable. There was a small allowance for additional tests.

The experiment was planned for a glove box that was installed and not yet "hot." The plan minimized costs associated with glove-box disposal. If the use of a new glove box were to become impractical, an existing inactive glove box would be coated with a strippable paint to provide a surface free of confounding contamination.

The study was to be conducted in collaboration with NMT-9 technical staff members at TA-55 and with the help of a primary NMT-9 collaborator and support and approval from the group leader. An uncontaminated glove box was to be identified. Assistance with the design of the experiment to ensure statistical relevance was to be sought from an appropriate staff member.

Actual. The actual method differs from the proposed method in a number of aspects, only a few of which are anticipated to significantly impact the experiment.

The new glove box was not available as anticipated. The glove box that will be used has had no contaminating operations performed in it; however there is contamination within it at levels of 10s to 100s of thousands of disintegrations per minute, which raises the potential for cross-contamination. Because the test samples are contained within covered petri dishes and the detector is kept in a ziplock bag, the effects of this contamination are expected to be minimal.

The complexity of using holding jigs that will not affect other samples has resulted in our using single samples in single petri dishes. The volume taken by individual petri dishes is considerably greater than originally planned; consequently, 14 petri dishes were made to limit operational impacts of the experiment, i.e., the number of dishes that fit in a holding rack. We judge that 14 will provide adequate results.

We have learned the air proportional probe we planned to use will not work in the glove box. Research has revealed that only 0.1% O₂ in a pure argon environment will result in erratic results in the proportional region of detector operation. The response in the glove box matches that observed in the laboratory and explained in the literature. As a result, we used a beta-gamma detector to obtain the first set of data. The beta-gamma background with this probe will greatly complicate statistical analysis of data obtained. We are investigating use of a sealed gas-proportional probe to solve these problems.

Progress and results

Overall progress has been disappointing. Numerous delays occurred in getting the project started, and consequently, only one set of data has been obtained. Furthermore, the data may not be useable. Because we base our results on a rate, one set of data does not produce a result. On the positive side: the significant portion of costs has been covered, the experiment has begun, and we will continue to gather and analyze data.

The deposition rate of ²³⁸Pu inside one glove box has been determined and may be useful for other studies.

Conclusion

The difficulties of working within a glove-box line were not fully recognized at the beginning of this project. We anticipated delays and setbacks but not to the extent we encountered them. We have begun the experiment and will continue the work.

Industrial Hygiene



*Reusability of Organic Vapor Air-
Purifying Respirator Cartridges*

*FRHAM-TEX II Cool Suit Material
Testing for Water (and Therefore,
Tritium) Protection*

*A Polymeric Barrier Monitor to Protect
Workers*

Reusability of Organic Vapor Air-Purifying Respirator Cartridges

Principal Investigators: Gerry Wood (PI), Richard Kissane (Col), Industrial Hygiene and Safety (ESH-5)

Funding: FY97, \$60K; FY98, \$61.1K; US Army (Supplemental Work), \$45K

Introduction

The purpose of this two-year study is to develop data to define the conditions, if any, under which an organic vapor (OV) air-purifying respirator cartridge can be reused. Two types of OV air-purifying respirator cartridges, MSA GMC-H and GME-H, are currently being used at Los Alamos National Laboratory. In the absence of information on their reusability, the best practice has been to use such cartridges only once before disposing of them. However, questions remain: Does one use mean use for only one work shift or does it mean use for one work period between breaks? Can a cartridge be used for one work week? Are there storage conditions that could extend the number of times a cartridge can be used?

Improved worker protection and cost savings are the potential benefits to the Laboratory from understanding the effects of usage breaks and storage conditions on the remaining service life of an OV cartridge. A tendency exists to reuse OV cartridges as a matter of convenience and for cost savings. In industrial operations, especially paint shops, excessive reuse of cartridges may be a serious problem. As we generate appropriate data and disseminate it at our Laboratory and to the industrial hygiene community, whatever the conclusion, the information will help in worker training and supervision and could reduce worker exposures to solvents and other OVs and gases. An obvious, though secondary, benefit of demonstrating the reusability of OV cartridges is cost savings. Depending upon the answers to our questions, users may need to purchase, store, distribute, and dispose of fewer cartridges.

The objectives of this work are to

- Develop experimental data on vapor migration within OV cartridges used at the Laboratory after single or multiple uses and storage under various conditions; and
- Prepare a mathematical model and computer program that provides guidance to industrial hygienists on the reusability of OV cartridges under actual use and storage conditions (vapor, concentration, humidity, temperature, times, etc.).

We will disseminate the data and interpretive models to the industrial hygiene community and the military, as well as to Laboratory industrial hygienists.

Method

We set up and calibrated an experimental apparatus for loading respirator cartridges with OVs and measuring vapor breakthrough before and after periods of storage. The cartridge study system was set up in a ventilation hood. A syringe pump was calibrated and used for generating vapors in flowing air for cartridge exposures. A breathing simulation pump generated cyclic airflow. We used a photoacoustic infrared analyzer to measure (at approximately one-minute intervals) the vapor concentrations exiting the test cartridges. The test system was automated for timed starts, stops, and analyses. Analyzer output was collected on a computer for further treatment. Cartridge storage was in sealed plastic bags at ambient temperatures and pressures.

Our usual procedure was to seal a freshly opened, weighed respirator cartridge into the airflow manifold between the point of vapor generation and the analyzer. The breathing pump and syringe pump were preset to deliver the desired average volumes of air (25 L/min) and liquid, respectively, to

give the desired vapor concentration (usually 1000 ppm). To begin an experiment the pumps and analyzer were started simultaneously. Any vapor breaking through the test cartridge was monitored with the analyzer for a simulated initial use period (0.5–2 hours). Then we removed the cartridge, reweighed it, and sealed it in a plastic bag for a selected time. After this storage period, we reinstalled the cartridge in the test manifold and rechallenged it with vapor at the same concentration. The vapor breakthrough was monitored to the point of cartridge saturation, where it reached a maximum. The final step was to again weigh the cartridge to determine the vapor saturation capacity at the test concentration.

Results

Our first accomplishment was to demonstrate that after a period of storage, there really is a problem with reusability of OV respirator cartridges. Figure 1 shows an example of this effect with ethyl acetate. The continuous breakthrough curve is the vapor effluent from a GMC-H cartridge for a continuous challenge of 1000 ppm for six hours. The other curve has a “step” change, which corresponds to the effect of 63 hours (long weekend) of storage without airflow after two hours of initial simulated use (at 990 ppm). The first five measurements after the storage period averaged 200 ppm as compared with an average of 2 ppm just before the end of the initial simulated use.

Ethyl acetate was the first vapor we used to study the effect of storage times and initial vapor loadings on reusability. Figure 2 shows averages of changes of vapor concentrations after various storage periods. The last five measurements before storage were averaged and subtracted from the average of the first

five after storage to get each change. The initial simulated uses were for two hours at 990-1026 ppm (upper curve) or one hour at 1008-1077 ppm. After four hours of storage with the higher loadings (longer initial use), there was a rapid increase in concentration of vapors released. For the lower loadings significant changes in simulated exposure occurred after 16 hours of storage and rose much more slowly.

Methylene chloride, a solvent commonly used at Los Alamos, was the second vapor we studied. It has a molecular weight similar to ethyl acetate but is more volatile. The Occupational Safety and Health Administration's permissible exposure limit was reduced from 500 ppm to 25 ppm on April 10, 1997, making the possibilities of excessive exposures on cartridge reuse more significant.

We studied the effect of methylene chloride concentration during the initial use period. Figure 3 shows two sets of experiments with about the same (30000 ppm-min) loadings, but at 1000 or 2000 ppm for 30 minutes or 15 minutes, respectively. Challenge vapor concentrations after storage were kept the same as before storage. The first observation is that the initial loading concentration did not affect the concentration changes up to 72 hours storage. Beyond that, the differences in the changes can be attributed to different challenge concentrations during the simulated reuse period. Second, significant vapor changes upon reuse occurred after only two hours of storage. Experiments at twice these loadings (i.e., about 2000 ppm for 30 min) also showed rapid (one hour), but higher storage changes (about 800 ppm at 24 hours).

Hexane was the third vapor we studied. Figure 4 shows the results for 0.5 or one hour simulated initial use of about 1000 ppm. Storage times lasted up to 14 days. In these cases, no changes in effluent vapor concentrations were observed within two or three days, respectively.

We also conducted some preliminary studies of effects of water vapor preloading

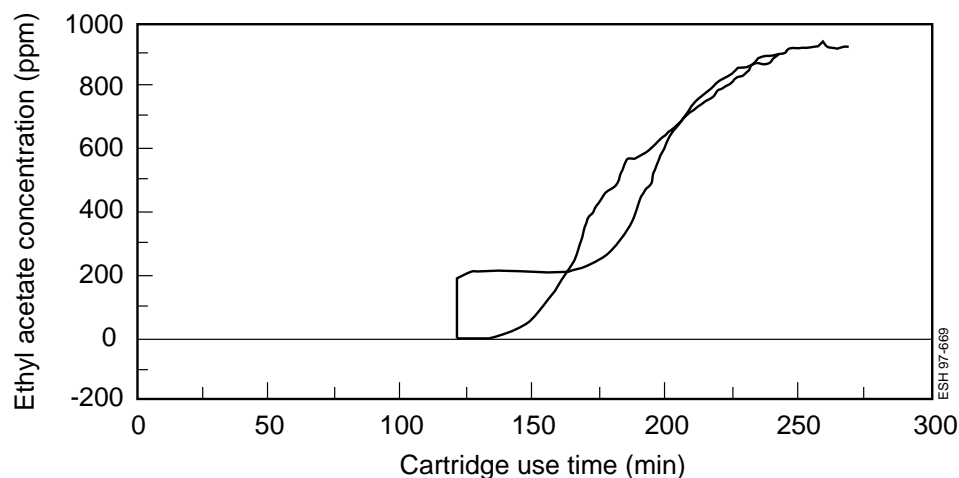


Figure 1. An example of the effect of storage on the concentration of vapor in the effluent with and without a storage period between uses. The challenges were about 1000-ppm ethyl acetate with a GMC-H cartridge. The storage time was 63 hours.

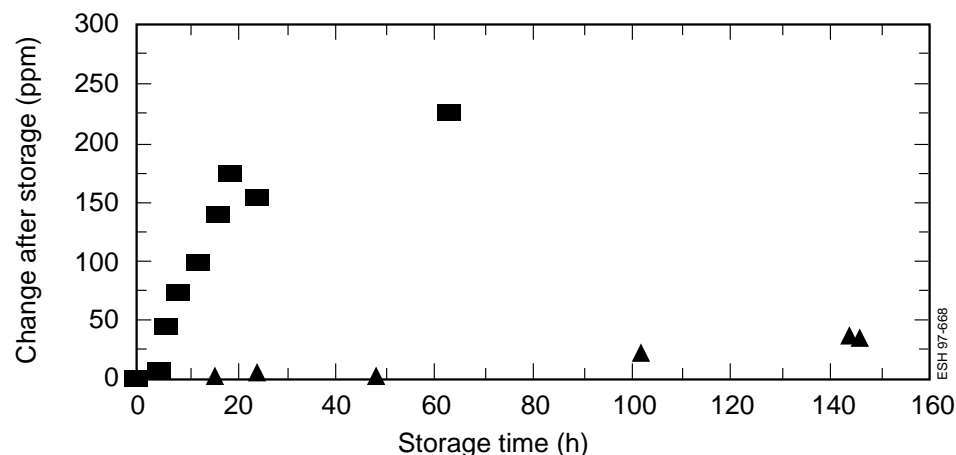


Figure 2. Ethyl acetate vapor. Effects of storage times and extents of loading on the changes in effluent vapor concentration. Squares = two-hour challenges at about 1000 ppm before and after storage. Triangles = one-hour challenges before storage.

and concurrent loading in humid air situations. We preloaded two cartridges with water by flowing humid (63–68% relative humidity) air through them for one hour before introducing the hexane vapor at 1000 ppm for one hour. Two other cartridges were simply run with 1000-ppm hexane in humid (53–55% relative humidity) air. After three days of storage the vapor concentration changes upon simulated reuse averaged 32 ppm for the preloaded cartridges and 11 ppm for the fresh cartridges run in humid air. These compare with an interpolated value of 4 ppm for the same vapor concentration in dry air (figure 4).

Conclusions

We have demonstrated increases in breathed vapor from simulated once-used cartridges upon reuse. This has been shown with three chemical vapors at a variety of conditions. After an initial period of storage the breathed vapor concentration changes increase with time of storage (figures 2-4). The type of vapor and extent of loading, but not so much the loading concentration (figure 3), affect the length of this initial period and the rate and extent of breathed vapor changes.

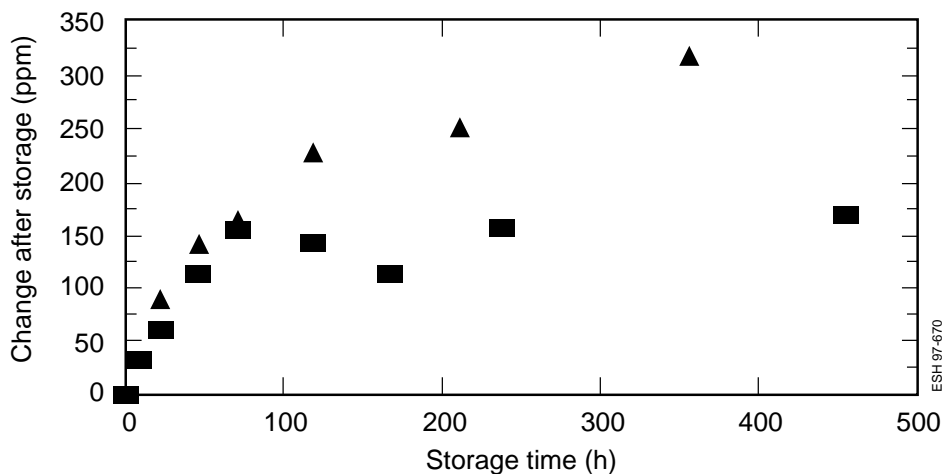


Figure 3. Methylene chloride vapor. Effects of storage times and challenge concentration on the changes in effluent vapor concentration. Squares = 30 minute challenges at about 1000 ppm before and after storage. Triangles = 15-minute challenges at about 2000 ppm. Both initial loadings were about the same.

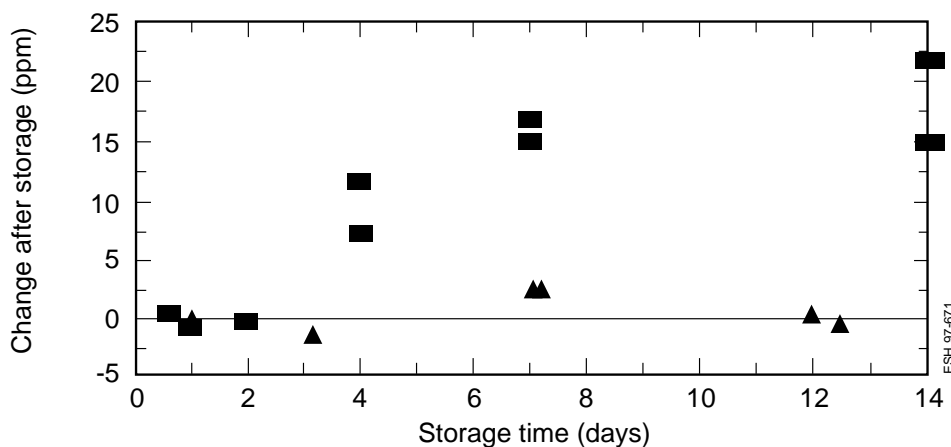


Figure 4. Hexane vapor. Effects of storage times and extents of loadings on the changes in effluent vapor concentration. Squares = one-hour challenges at about 1000 ppm before and after storage. Triangles = 0.5 hour challenges at the same conditions.

The vapor effluent changes upon storage were greatest for methylene chloride, the most volatile chemical, and least for hexane, the least volatile.

Water vapor coadsorption (hexane vapor in humid air exposure) resulted in higher storage effects for three days of storage. Water vapor preadsorption (humid air exposure before and during hexane vapor challenge) resulted in even higher effects.

Qualitatively, the observed effects of loading during the initial cartridge use, storage time, vapor type, and water vapor are what we expected. However, what

we now have are quantitative measurements of these effects for three different chemical vapors. With the data accumulated, we can begin to model the adsorbed vapor migration, which will be the next phase of the study. Then we can begin to investigate these effects at other storage conditions that may mitigate them.

FRHAM-TEX II Cool Suit Material Testing for Water (and Therefore, Tritium) Protection

Principal Investigators: Gerry Wood (PI), Richard Kissane (Col), Industrial Hygiene and Safety Group (ESH-5)

Funding: FY97, \$64.6K; Hanford, \$9K

Introduction

We studied FRHAM-TEX II, a new clothing material, to determine its effectiveness in protecting workers against water and water vapor, either of which may be tritiated. FRHAM-TEX II in a cool suit configuration is claimed by the manufacturer to provide more comfort than other materials because of its ability to transpire liquids, i.e., remove sweat. The manufacturer also intends that their cool suit protect workers by repelling water and chemicals. Cool suits (with caps) made of FRHAM-TEX II are being offered to workers at Los Alamos National Laboratory as an option to their use of Tyvek clothing.

As a result of this study, we intended to provide supervisors, users, and purchasing agents with guidance concerning the effectiveness of FRHAM-TEX II clothing for water repellence. Based on objective data rather than on manufacturers' claims or personal preference, potential users could then select protective clothing in place of or in addition to Tyvek clothing. Additionally, cost savings might be realized by potential users' having a choice among protective clothing materials that are shown to be equally effective, whether they are FRHAM-TEX II, Tyvek, or another material. Improved worker protection may also result from the users' ability to select better materials for the workplace.

After our work was underway, we received funding from Hanford to study six samples of protective clothing they were considering using. They requested that, among other properties, we study water repellence. Hanford's interest provided us with more materials to compare to FRHAM-TEX II and demonstrated another benefit of our study—having a procedure and equipment

developed and available for future studies of this type, including studies of liquid and vapor organic chemicals.

Method

The materials we used in these studies were cut from yellow or magenta FRHAM-TEX II protective clothing articles in the Laboratory's inventory, from a white sample of FRHAM-TEX II provided by the manufacturer, or from six brands of suits sent to us from Hanford. Normal (untritiated) water or water vapor challenged the test material.

To conduct water permeation measurements, we mounted fresh or dried samples in a standard permeation cell with either the outside (usually) or the inside on the challenge side of the cell. The permeation cell has one or more necks on the challenge side into which water can be poured or through which humidified air can be passed. The measurement side of the cell has one neck for a thermocouple and two more for air circulation. The cell is designed so that dry, sweep air impinges on the measurement side of the test material and swirls around before exiting to the analyzer. The area of material exposed to challenge water (or water vapor) and sweep air is 25 cm².

The water vapor analyzer we used is an EG&G Model-911 Dew Point Hygrometer. We measured the dew point or frost point and converted these measurements to water concentrations (mg/L) using the ideal gas law and correlations based on tabulated vapor pressures. We multiplied the air sweep rate (L/min) by the increase of water vapor concentration upon water or water vapor challenge to derive the water vapor permeation rate (mg/min).

We regulated and calibrated dry air from a cylinder to a 1.0 L/min flow rate

through the test cell and recorded on a strip chart the analog output of the measured dew/frost point. At selected times, we also manually recorded digital dew/frost point and/or relative humidity (RH) readouts from the analyzer. A baseline, usually with less than 0.002°C/min frost point drift, was established before a test began. This corresponded to 3–5% RH of the sweep air. The initial frost point and cell temperature were recorded. We then quickly poured water into the challenge side of the cell or exposed it to a 1.0 L/min flow of humid air. In the latter case, we passed compressed air over a water bath, heated if necessary, to give the desired challenge water vapor concentration. Just before beginning the test, we monitored the dew point of the humid air until it stabilized.

We also conducted tests at above-ambient temperatures. We heated the cell with a voltage-controlled heating tape before and during the test and monitored temperature.

Two liquid water splash tests were done with a FRHAM-TEX II sample (outside contact). We rapidly poured 5 mL of water into the challenge side of the cell, immediately poured it out, and then inverted the cell to drain. We continued measurements to a maximum value and through the subsequent decrease.

Results

One of the Hanford-supplied materials, Copiah Creek, immediately soaked up and passed liquid water through it; therefore, we discontinued using it for any further liquid water tests. All of the other materials repelled liquid water—we observed no drops on the other side—even though in most cases, vapor permeation was measured.

Table 1 shows a summary of the results of the liquid water challenge tests. Except for the Kappler Tyvek material, water permeation started within 10 seconds of adding the liquid to the challenge side. The vapor concentration increased so fast that the analyzer overshoot and undershot the steady-state value several times until the signal leveled off, usually within five minutes. For the splash tests, table 1 presents the maximum permeation rates, rather than steady rates.

The best correlation we found for the effect of water vapor concentration on water vapor permeation is shown in figure 1. The "critical" water vapor concentration of about 12 mg/L at 20°C corresponds to 70% RH.

Table 2 shows a summary of the results of the higher water vapor challenge tests. Over the 65–75%RH range, we did not see a significant difference between permeation from the inside and the outside of the FRHAM-TEX II material. However, the permeation from the inside seems to be significantly higher than from the outside at the highest humidities. This may be the transpiration effect claimed by the manufacturer. However, the LANL Tyvek also showed this effect.

We also studied the temperature effect on permeation from the outside of FRHAM-TEX II material. We conducted a series of experiments in which water vapor challenge concentration was kept constant at 15.5 ± 0.3 mg/L, while the cell temperature varied from 17–37°C. Figure 2 presents the results. Except at the lowest temperatures (17–19°C), permeation rates were essentially the same. We suspect that the highest rates in both figures 1 and 2 are due to liquid water condensation on the challenge surfaces at the lower temperatures involved.

Table 1. Water permeation for liquid water challenges

Material	Challenge side	Cell temperature (°C)	Number of tests	Water permeation Rate (mg/min) average	range
FRHAM-TEX II	Outside	23	9	8.4	$\pm 0.7^*$
FRHAM-TEX II	Inside	24	2	8.5	± 0.4
FRHAM-TEX II	Outside	36	2	11.2	± 0.1
FRHAM-TEX II	Outside**	23	2	3.2	± 0.2
LANL Tyvek	Outside	24	2	5.4	± 0.3
LANL Tyvek	Inside	24	2	4.7	± 0.2
TSO-150	Outside	24	2	5.1	± 0.3
Kappler ProShield 2	Outside	24	2	4.7	± 0.1
Kappler NUFAB	Outside	24	2	7.0	± 0.4
Kool Cool Suit	Outside	24	2	1.2	± 0.0
Kappler Tyvek	Outside	24	2	0.0	± 0.0

* Standard deviation estimate

** Splash tests (maximum permeation rates)

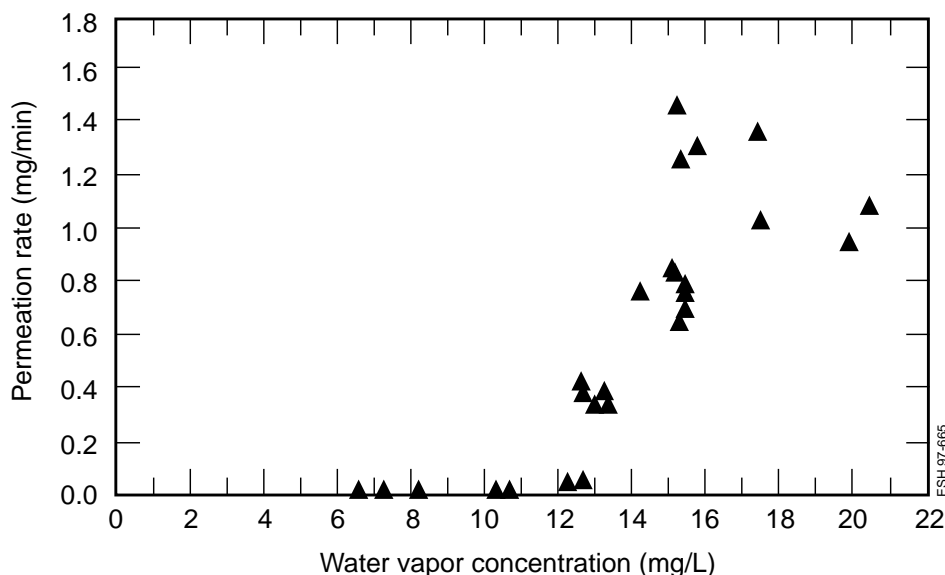


Figure 1. Effect of water vapor concentration on water vapor permeation of FRHAM-TEXII material. All challenges were to the outside of the material at $20 \pm 3^\circ \text{C}$.

Table 2. Water permeation for higher water vapor challenges

Material	Challenge side	Challenge humidity range	Cell temperature (°C)	Number of tests	Water permeation Rate (mg/min)	
					average	range
FRHAM-TEX II	Outside	65–75% RH	21	3	0.38	±0.03
FRHAM-TEX II	Inside	65–75% RH	21	3	0.42	±0.02
FRHAM-TEX II	Outside	> 93% RH	22	4	1.1	±0.2
FRHAM-TEX II	Inside	> 93% RH	23	4	1.9	±0.3
LANL Tyvek	Outside	> 98% RH	22	2	3.15	±0.03
LANL Tyvek	Inside	> 98% RH	23	2	4.07	±0.01
Kappler Tyvek	Outside	> 98% RH	23	2	Not detected	
Copiah Creek	Outside	97% RH	25	1	6.2	
FRHAM-TEX II (white)	Outside	78–82% RH	25	5	0.7	±0.4

Conclusions

Water repellence can be defined as the inverse of water permeation rate upon liquid water exposure. Of the materials studied, we found repellence to be the best for the Kappler Tyvek material and the worse for the Copiah Creek material. The latter was apparently an uncoated, woven cloth. FRHAM-TEX II and LANL Tyvek—the type currently used at Los Alamos—permeation rates fell between these extremes for both liquid water and water vapor challenges. Of these two Los Alamos samples, Tyvek was a little better for water repellence, but worse for water vapor permeation at high humidities. When exposed to liquid water, neither material is a very good water vapor barrier.

We saw no big differences between liquid water repellence for the two directions of permeation for FRHAM-TEX II or Tyvek. However, significant differences did appear for the highest humidity vapor permeation studies.

Temperature effects on vapor permeation occurred only at the lowest temperatures, when condensation on the outside surface could explain higher rates.

Splash tests showed that immediate water permeation occurs for even the briefest exposure of FRHAM-TEX II to liquid water. This was confirmed by the

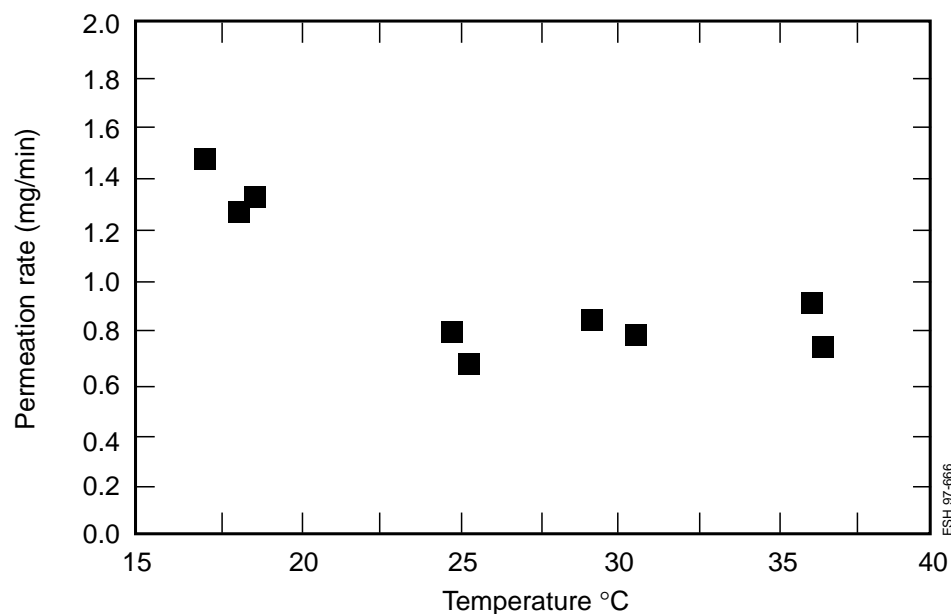


Figure 2. Effect of temperature on the water vapor permeation of FRHAM-TEX II material. All challenges to the outside of the material at 15.5 ±0.3-mg/L water vapor concentration.

rapid permeations observed for the constant water exposure experiments.

Figure 1 shows that for FRHAM-TEX II material, water vapor permeation increases rapidly above 12 mg/L vapor concentration (about 70% RH) at 20°C. Therefore, FRHAM-TEX II is not a good barrier for water vapor at high humidities.

A Polymeric Barrier Monitor to Protect Workers

Principal Investigators: R. Hermes, Material Technology/Polymers and Coatings (MST-7); T. Stampfer, visiting scientist, Industrial Hygiene and Safety Group (ESH-5)

Funding Level: FY97, \$49K; \$25K returned to TDEA July 1997.

Introduction

The original project to develop gloves made of a material that offered a polymeric barrier monitor to protect workers began in FY95 and has continued through FY97. The principal investigators established the objective—to fabricate a new type of glove that with proper electrical connections could sound an alarm when breached by a puncture from sharp objects made of metal, glass, or wood.

The new glove's utility would be in its replacing the current lead-lined glove in glove-box operations dealing with radioactivity, such as those used at the Plutonium Facility, the Chemistry Metallurgy Research facility, etc. The outcome we sought was that an instant alarm would occur when the integrity of a glove was breached during use. The benefit would be for radiological worker protection in real time.

Our goal for FY97 was to study the gloves delivered at the end of FY96 and to investigate why they did not perform according to expectations, correct the problem(s), and have new gloves ready to test. We defined the following tasks:

On an iterative basis, purchase multilayered film slabs from North Hand Protection, Charleston, South Carolina, with various formulations for improving processability and raising the electrical conductivity.

Having established the proper formulation, purchase hand-length gloves having the pentalayered structure, the original design feature from FY95.

Test the gloves for penetration detection capability using the appropriate electrical circuit and puncturing devices.

Progress and results

Several iterations of material were purchased on a regular basis from North Hand Protection. The initial formulations attempted in FY96—carbon-filled butyl rubber—were tried again, but with a different type of conducting carbon. Although we were able to achieve reasonable conductivities with the newly formulated film slabs, the result was a reduction in processability. More carbon loading led to higher conductivity but poorer film quality (ripples and separation from the base coat of Hypalon™).

Problems associated with the carbon-filled butyl rubber prompted us to change to carbon-loaded Neoprene™ in June, the ninth month of the project. In mid-August, we received several samples which we used to evaluate the electrical properties of the butyl and Neoprene™ materials. The data are shown in table 1.

The lower the surface resistivity and the lower the point-to-point resistance, the better was the result. Processability of the regular Neoprene filled at a level of 40% conductive carbon afforded the best result.

Unfortunately, our development work was directly tied to the production schedule at North Hand Production; that is, the preparation of our samples depended upon their ability to obtain the raw materials and to fit our requests into their production schedules. Consequently, spending on the project by mid-year indicated to us that we might not be able to use the entire FY97 appropriation from TDEA. We then made arrangements to return more than half the appropriation to the TDEA account.

Deliverables

- During this project we attained one of our deliverables—a material with a reasonable balance between conductivity and processability.

- Because we could not get gloves made by the end of the fiscal year, we turned the project over to the Actinide Process Chemistry Group (NMT-2). Although their FY98 budget is limited, they plan to continue the project.

Table 1. Test data for film slab formulations

Sample	Description	Surface resistivity (ohms/sq)	Resistance (ohms)	Processability
C-30	butyl/30phr carbon	4 X 10 ³	6600	poor
C-40	butyl/40phr carbon	1 X 10 ³	2200	poor
NFB-25	soft Neoprene/30phr carbon	3.5 X 10 ⁶	>35 M	poor
NFB-50	soft Neoprene/50phr carbon	1 X 10 ³	2400	very poor
NGW-40	reg. Neoprene/40phr carbon	3 X 10 ³	20000	very good

Publications and Presentations

The publications and presentations listed below have received full or partial funding from the TDEA program. In some instances a project has been ongoing and TDEA contributed toward supporting progress.

Fresquez et al.

Biggs, J., K. Bennett, and P. R. Fresquez, "Application of Global Positioning System (GPS) Radio Collars to Rocky Mountain Elk Studies at Los Alamos National Laboratory," LAUR-96-3393, Los Alamos National Laboratory, 1996.

Biggs, J., K. Bennett, and P. R. Fresquez, "Evaluation of Habitat Use by Rocky Mountain Elk in Northcentral New Mexico Using Global Positioning System (GPS) Radio Collars," submitted to *Journal of Wildlife Management* June 1997.

Bennett, K., J. Biggs, and P. R. Fresquez, "Accuracy Determination of Global Positioning System (GPS) Radio Collars in Relation to Vegetation Canopy and Topographical Influences of Northcentral New Mexico," submitted to *Journal of Wildlife Management* June 1997.

Bennett, K., J. Biggs, and P. R. Fresquez, "Determination of Locational Error Associated with GPS Radio Collars in Relation to Vegetation Canopy and Topographical Influences of Northcentral New Mexico," LA-UR-96-4921, Los Alamos National Laboratory, 1996.

Biggs, James, Kathryn Bennett, P.R. Fresquez, and R. Robinson, "Movements, Disease Analysis, and Tritium Concentrations of Rocky Mountain Elk of the Pajarito Plateau," LA-UR-3395, Los Alamos National Laboratory, 1996.

Biggs, J., K. Bennett, and P. R. Fresquez, "Estimation of Home Ranges and Water Use of Elk at Los Alamos National Laboratory," LA-UR-97-3977, Los Alamos National Laboratory, 1997.

A Symposium of Biological Research in the Jemez Mountains, New Mexico, Santa Fe, NM, October 18, 1996:

"Accuracy Determination of GPS-borne radio collars in Relation to Vegetation Canopy and Topographical Influences of Northcentral New Mexico," K. Bennett, J. Biggs, and P. R. Fresquez.

"Application of GPS-borne Radio Collars to Rocky Mountain Elk Studies at Los Alamos National Laboratory," J. Biggs, K. Bennett, and P. R. Fresquez.

"Movements, Disease Analysis, and Tritium Concentrations of Rocky Mountain Elk of the Pajarito Plateau," J. Biggs, K. Bennett, P. R. Fresquez and R. Robinson.

The Wildlife Society Joint Meeting—AZ/NM, Gallup, NM February 1997:

"Determination of Locational Error Associated with (GPS) Radio Collars in Relation to Vegetation Canopy and Topographical Influences of Northcentral New Mexico," K. Bennett, J. Biggs, and P. R. Fresquez.

"Evaluation of Habitat Use by Rocky Mountain Elk in Northcentral New Mexico Using Global Positioning System (GPS) Radio Collars," J. Biggs and K. Bennett.

The Wildlife Society Meeting, Snowmass, CO, September 1997:

"Interfacing of GIS/GPS radio-collar data to model large mammal movements," K. Bennett and J. Biggs.

"Evaluation of habitat use by Rocky Mountain elk in north central New Mexico," J. Biggs, K. Bennett, and P. R. Fresquez.

"Participation in conference workshop for 'advanced telemetry systems in wildlife studies' addressing locational error associated with GPS radio collars," K. Bennett and J. Biggs.

Ecological Society of America Annual Meeting, Albuquerque, August 1997:

"Modeling distribution and activity patterns of Rocky Mountain elk using global positioning system radio collars and the geographical information system," J. Biggs, K. Bennett, and P. R. Fresquez.

Devine et al.

Hsu, H-H. and Devine, R. T., "Neutron Charged Particle Reactions in CR-39 Using MCNP and LAHET," *Health Physics of Radiation-Generating Machines, Proceedings of the 30th Midyear Topical Meeting*, 5–8 January 1997, San Jose, California, p. 181.

Walker, L. S. et al., "Response of a Health Physics Instrument Model 2080 Albatross to a High- Energy Neutron Field," *Health Physics of Radiation-Generating Machines, Proceedings of the 30th Midyear Topical Meeting*, 5–8 January 1997, San Jose, California, p. 163.

Walker, L. S. et al., "Trouble Shooting, Modification, and Calibration of a Bismuth Fission Chamber Used in Conjunction with Bonner Spheres to Measure High-Energy Neutron Flux," *Health Physics of Radiation-Generating Machines, Proceedings of the 30th Midyear Topical Meeting*, 5–8 January 1997, San Jose, California, p. 235.

Wender, S.A., Walker, L.S., and Ullmann, J.L., "Capabilities of the WNR High-Energy Neutron Beam at LANSCE for High-Energy Neutron Health Physics Research," *Health Physics of Radiation-Generating Machines, Proceedings of the 30th Midyear Topical Meeting*, 5–8 January 1997, San Jose, California, p. 487.

Duran, M., Casson, W., and Walker, L. S., "High-Energy Spectral Measurements. THAM-C.5," presented at the 45th Annual Meeting of the Health Physics Society, 29 June–3 July 1997, San Antonio, Texas.

Hansen et al.

We presented the results of the first year of this project at the following meetings:

Hansen, L. A., R. J. Robinson, P. R. Fresquez, T. S. Foxx, and J. D. Huchton. "Evaluation of radio frequency identification technology to monitor medium-sized mammal use of potential contaminant release sites," Fourth Wildlife Society Conference, 21–27 September, 1997, Snowmass, Colorado (poster).

Hansen, L. A., R. J. Robinson, P. R. Fresquez, T. S. Foxx, and J. D. Huchton. "Movements of and contaminants in medium-sized mammals around a liquid waste lagoon at Los Alamos National Laboratory," Second Symposium of Biological Research in the Jemez Mountains, 7 November, 1997, Santa Fe, New Mexico (presentation).

Hermes et al.

The glove material patent application is filed under LANL file #S-84,971.

After several iterations between the US Patent Office and the Laboratory's patent attorney, the new glove material was awarded a "notice of allowable subject matter." The Laboratory paid an issue fee in August 1997. The patent application is now categorized "patent pending," which means the patent will be issued.

Inkret et al.

Results presented at 1996 Annual Meeting of Health Physics Society and the Annual Meeting on Bioassay, and Environmental Radiochemistry. Manuscripts are in preparation for publication in the open literature.

Mallett et al.

Mallett, M. W., "Neutron extremity dosimetry at LANL," *Proceedings of the 1996 Harshaw-NE Users Group Symposium on External Dosimetry: Integrating Technologies and Methods*, 1996.

Wood-Zika, A. R., "Calculation of Extremity Neutron Fluence-to-Dose Equivalent Conversion Factors," Texas A&M University, M.S. thesis 1997.

Rees et al.

The lessons learned while introducing a probe to the glove-box line will be written up and submitted for presentation to the American Glovebox Society's 1998 meeting.

We plan to publish our results about resuspension.

Rodgers et al.

McFarland, A., J.C. Rodgers, and M.J. Koskelo, "Sampling alpha-emitting transuranic aerosols in the nuclear workplace," Los Alamos National Laboratory Report LA-UR-97-3646 (1997).

Rodgers, J.C., J.J. Whicker, and J.T. Voss, "Comparison of continuous air monitor utilization: A case study," Los Alamos National Laboratory Report LAUR-97-1345, Submitted to Radiation Protection Management (1997).

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